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ALPHA-GAMMA ANGULAR CORRELATION MEASUREMENTS WITH LIQUID SOURCES

by

Elias Smith Murphy, Jr.

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ABSTRACT

Alpha-gamma angular correlation measurements have been made with solid sources of Am^{243} and with liquid sources containing either Am^{243} or an even-even alpha emitter in dilute perchloric acid solutions. Even-even alpha emitters which have been studied are U^{232}, Th^{230}, and Ra^{226}. Thicknesses of the solid sources were controlled so that the neptunium recoils from one source were stopped in Am_2O_3, while recoils from the other sources were stopped in the aluminum, gold, or mica backing on which the sources were vaporized. The liquid sources were films consisting of 3 microliters of solution placed between a rubber hydrochloride membrane and a microscope cover glass, 1 cm^2 in circular cross section. The perchloric acid concentration of the liquid sources ranged from 0.5 to 3.0 molar. All of the angular correlations obtained with solid Am^{243} sources were attenuated, the average attenuation coefficients being 0.29 ± 0.01 for sources in which recoils were stopped in Am_2O_3, 0.20 ± 0.01 for sources in which recoils were stopped in mica, 0.52 ± 0.02 for sources in which recoils were stopped in gold, and 0.67 ± 0.01 for sources in which recoils were stopped in aluminum. Unattenuated angular correlations were obtained with liquid sources containing Am^{243} in 0.5 M and 1.0 M $HClO_4$. For liquid sources containing Am^{243} in 3.0 M $HClO_4$, the correlation was attenuated, with an average attenuation coefficient of 0.86 ± 0.01. Attenuated angular correlations were also found with liquid sources containing an even-even nuclide in dilute aqueous solutions. The average attenuation coefficients for the even-even nuclide liquid sources were $G_2 = 0.75 \pm 0.05$ and $G_4 = 0.86 \pm 0.03$ for U^{232}, $G_2 = 0.64 \pm 0.06$ and $G_4 = 0.84 \pm 0.04$ for Th^{230}, and $G_2 = 0.76 \pm 0.11$ and $G_4 = 0.90 \pm 0.07$ for Ra^{226}. The postulate that the measured angular correlation function for Am^{243} is attenuated by the same amount as the correlation functions for the even-even nuclides is found to be untenable since it requires unreasonable alpha mixing ratios or unreasonable admixtures of M2 to E1 gamma radiation in the Am^{243} decay process. The attenuation observed with even-even nuclide sources is believed to be consistent with the postulate of a time-dependent electric interaction.

I. INTRODUCTION

A. Angular Correlation Measurements

In an appreciable fraction of alpha decays, emission of the alpha particle results in an excited level of the daughter nucleus which then decays to the ground level by emission of a gamma photon. For an ensemble of radioactive nuclei, the alpha-gamma angular correlation is defined as the angular distribution in the intensity of the gamma radiation de-exciting an intermediate level, measured with respect to the direction of propagation of the alpha radiation which led to formation of the level.

The intensity distribution of gamma radiation from excited nuclei depends upon the multipolarity of the radiation and upon the angle between the direction of emission of the radiation and the nuclear spin axis. At ordinary temperatures and in the absence of strong magnetic fields, the angular distribution of gamma radiation from a sample containing many nuclei is isotropic because the nuclei in the sample have their spins oriented in a random manner. In order to obtain an anisotropic intensity pattern, it is necessary to observe gamma radiation from nuclei whose spins are preferentially oriented (i.e., from nuclei for which there is an unequal population of the magnetic sublevels of the emitting state). One method for obtaining the necessary spin orientation is to cool the sample to a very low temperature while at the same time subjecting the sample to a strong magnetic field. An alternative method, for the case of gamma radiation which follows alpha decay, is to consider only photons which follow alpha particles emitted in a preassigned direction. The orientation of the angular momentum of the intermediate level following alpha decay is determined by the multipolarity of the alpha particle and its direction of emission. If this intermediate level does not have spin 0 or $\frac{1}{2}$, its magnetic sublevel population relative to the direction of alpha-particle emission as the axis of quantization will be unequally distributed. Then, if the lifetime of the intermediate level is short enough so that the magnetic sublevel population is not altered, the gamma radiation which de-excites this level will have an anisotropic intensity distribution.

The angular correlation can be expressed mathematically in the form

$$W(\theta) = 1 + \sum_{n} A_n P_n(\cos\theta) \quad ; \quad n = 2,4,\ldots \quad , \tag{1}$$

where $W(\theta)$ represents the relative probability of emission of gamma radiation at an angle θ with respect to an arbitrary direction of alpha emission. The $P_n(\cos\theta)$ are Legendre polynomials, and the A_n are constants which depend upon the spins of the nuclear levels involved in the

radioactive transition and upon the angular momenta carried away by the radiations. The angular correlation is often expressed in terms of the anisotropy, A, which is defined as

$$A = \frac{W(180°)}{W(90°)} - 1 \quad . \tag{2}$$

A zero value of A is ambiguous in that it can mean either that the correlation is isotropic or that (as in the case of the 0-2-0 alpha-gamma transition in even-even nuclei) $W(180°) = W(90°)$.

Many excellent review articles on angular correlation measurements have appeared in the literature.[1-4] The general theory of angular correlations was first developed by Hamilton.[5] The currently most widely used formulation of the theory is due to Biedenharn and Rose.[6] Theoretical values of A_n for the alpha-gamma transitions investigated in the present work are given in Chapter II.

B. Attenuation of the Correlation

Since the intensity distribution of gamma radiation is a function of the angle between the direction of emission of a photon and the nuclear spin axis, the possibility of observing an unperturbed distribution depends upon the magnetic sublevel population of the intermediate nuclear level remaining fixed during the intermediate level lifetime. In practice, for lifetimes in excess of a fraction of a millimicrosecond, interactions between the nucleus and its environment cause precession of the spin of the intermediate level resulting in a change in the magnetic sublevel population. This in turn causes the angular correlation to be attenuated. Interactions which cause precession of the nuclear spin can occur between the nuclear magnetic dipole moment and extranuclear magnetic fields, and between the nuclear electric quadrupole moment and extranuclear electric field gradients. These interactions may be either static, as in the case of an interaction between a nucleus and an externally applied magnetic field of constant magnitude and direction, or time dependent, as in the case of an interaction with varying (due to Brownian motion) electric field gradients in a liquid. For a static interaction, the correlation will be measurably attenuated if $\omega\tau_N \geq 0.1$, where ω is the precession frequency and τ_N is the lifetime of the intermediate level. For a time-dependent interaction, the correlation is subject to attenuation of the form $e^{-\lambda t}$, where λ depends both on the strength of the interaction and on the correlation time* of the perturbing electric or magnetic field.

*The "correlation time" is qualitatively defined as the time during which a local field configuration is preserved without undergoing an appreciable change.

There are two factors peculiar to alpha-gamma angular correlations which contribute to attenuation of the correlation. First of all, through the Migdal effect,[7] interactions between an ejected alpha particle and atomic electrons can leave an atom in an excited or an ionized state, resulting in electric and magnetic fields at the nucleus. Secondly, the nuclear recoil complicates the picture. As shown in Appendix I, the neptunium daughter from alpha decay of Am^{243} recoils with an energy of approximately 88 kev and may traverse 10-50 $\mu g/cm^2$ of matter before coming to rest in about 10^{-13} sec. Electrons can be stripped from the daughter atom or raised to excited states during recoil. When the recoil comes to rest, it is likely to occupy an intermediate position in the lattice, where the nucleus will be subject to electric field gradients. A recoiling atom loses energy both as a result of ionization and of elastic collisions with other atoms in its path. Ionization or displacement of the atoms in a crystal lattice may also give rise to electric field gradients at the point where the recoil finally comes to rest.

For an attenuated correlation, Eq. (1) becomes

$$W(\theta) = 1 + \sum_n G_n A_n P_n(\cos\theta) \quad ; \quad n = 2,4,\ldots \quad , \tag{3}$$

where the G_n are the attenuation coefficients. The dependence of G_n on the properties of the nucleus and on the magnitudes of external electric or magnetic fields is discussed in Chapter II.

C. The Problem

In the present investigation, alpha-gamma angular correlation measurements have been carried out with solid sources of Am^{243} and with liquid sources containing either Am^{243} or an even-even alpha emitter in dilute perchloric acid solutions. The even-even alpha emitters which have been studied are U^{232}, Th^{230}, and Ra^{226}. The perchloric acid concentration of the liquid sources ranged from 0.5 to 3.0 molar. For the solid sources, the thickness of the active layer was controlled so that for one source the neptunium recoils would be stopped in Am_2O_3, whereas for the other sources the recoils would be stopped in the gold, aluminum, or mica backing material.

For radioactive transitions for which the ground state spins of both parent and daughter nuclei are known, angular correlation measurements yield information about spins and parities of intermediate levels. In addition, alpha-gamma angular correlation measurements can be useful in providing fundamental data relating to the process of alpha emission. An alpha-gamma correlation measurement can provide information about the relative intensities and phases of the alpha waves of different multipolarities which participate in the transition being investigated. In order to obtain maximum information from alpha-gamma angular correlation

measurements, however, it must either be possible to observe the unperturbed correlation, or else the magnitude of the perturbation must be accurately known. The aim of the liquid source measurements which are reported here was to investigate the possibility of obtaining the unperturbed angular correlation with these sources. The solid source measurements on Am^{243} were performed in order to provide data for comparison with the results of the liquid source measurements, and also to obtain evidence to confirm results reported recently by Flamm.[8]

Most alpha-gamma angular correlation studies reported in the literature have been made with solid sources on even-even nuclei. The alpha-gamma transition which has been most extensively studied has been the one passing through the first excited state of the daughter nucleus (the 0-2-0 transition) for which the theoretical correlation function is well known. Invariably, these solid source measurements have resulted in attenuated correlations.

Since interactions between the nucleus and extranuclear electric and magnetic fields cause the angular correlation to be perturbed, it would seem logical to investigate the correlation as a function of the medium in which the nucleus is embedded. For gamma-gamma angular correlation experiments in which the nuclear environment was changed,[9,10] it was found that the correlation was less attenuated in a metallic environment than in a nonconducting environment. Flamm[8] has observed the same result with respect to perturbation of the alpha-gamma angular correlation. Gamma-gamma angular correlation measurements made on liquid sources showed that, in general, the correlation is less attenuated in liquid sources than it is in solid sources.[10-12] The only applications of liquid sources to alpha-gamma measurements so far reported have been those of Novey[13] and of Krohn, Novey, and Raboy[14] for Am^{241}.

Americium-243 was chosen as a nuclide to be investigated because (1) the half-life of its first excited state is long enough so that perturbation of the correlation function would be expected, (2) it has a high specific activity and a relatively high gamma/alpha ratio so that a statistically meaningful number of coincidences can be obtained in a few hours of counting time, and (3) other work on isotopes of americium has been reported so that data exist with which to compare the present measurements. Because the relative intensity of alpha-particle waves of multipolarity I = 0 and I = 2 which lead to the first excited state in Np^{239} can only be approximately determined, the theoretical correlation function for Am^{243} is not known with the certainty which is desirable. On the other hand, because the ground state spins of even-even nuclei are known to be zero, and because only alpha waves of a single multipolarity can arise in a transition from a zero spin state, the theoretical alpha-gamma angular correlation function for these nuclei can be determined unambiguously. In a situation where one would like to know if the unperturbed

correlation has been observed, it thus becomes desirable to investigate even-even nuclei. The principal difficulties with even-even nuclei are that (1) alpha decay is predominantly to the ground level of the daughter nuclide, and (2) gamma emission from the first excited level is very highly converted resulting in an unfavorable gamma/alpha ratio. Long counting times are thus required in order to obtain statistically meaningful coincidence rates.

II. REVIEW OF LITERATURE

A. Nuclear Properties of Am^{243}

A decay scheme diagram for the decay of Am^{243} to Np^{239} is shown in Fig. 1. The decay scheme is based on data reported by Stephens et al.,[15] Hummel,[16] and Strominger et al.[17] The half-life of the 74.6-kev level in Np^{239}, as measured by the delayed coincidence technique,[18] is $(1.2 \pm 0.1) \times 10^{-9}$ sec. Gamma radiation which depopulates the 74.6-kev level is believed to be E1 on the basis of internal conversion coefficient measurements.[19] The ground state spins of Am^{243} and Np^{239} are known to be 5/2 from measurements of the hyperfine structure of these isotopes.[20,21]

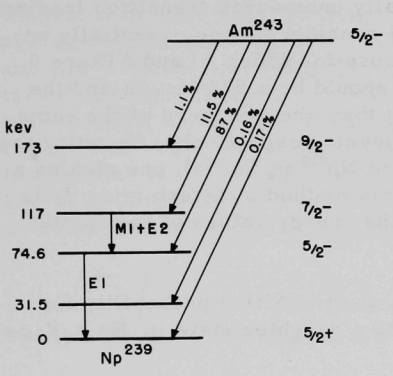

Fig. 1. Decay-scheme Diagram for the Alpha Decay of Am^{243} to Np^{239}.

Assignment of spin 5/2 to the 74.6-kev level in Np^{239} is based on consideration of the properties of the energy levels in rotational bands according to the collective nuclear model of Bohr and Mottelson.[22-24] Bohr and Mottelson picture the nucleus, in regions between closed shells, as an ellipsoidal body subject to deformations in the form of surface waves involving the collective motion of many nucleons. The various surface wave patterns represent quantized energy states characterized by quantum numbers I and K, where I is the nuclear angular momentum quantum number and K represents the projection of the total angular momentum on the nuclear symmetry axis. A rotational band contains levels which all have the same K. The energies of the levels in a rotational band are given by

$$E_I = \frac{\hbar^2}{2\Im}\left[I(I+1) - I_0(I_0+1)\right]; \qquad I_0 \neq 1/2 \quad , \tag{4}$$

where I_0 is the spin of the ground level, I is the spin of an excited level, and $\Im$ is the moment of inertia of the deformed nucleus. In an odd-A nucleus, the levels in a rotational band have angular momenta given by $I_0 = K$ and $I = K + 1, K + 2, \ldots$. For even-even nuclei, the angular momenta of a rotational set follow the sequence 0+, 2+, 4+, 6+....

In even-even nuclei it has been found that only alpha transitions to the ground state obey simple barrier-penetration theory. For alpha transitions to excited states, the observed partial half-life is always greater

than that predicted by theory, and these transitions are said to be hindered. The hindrance factor is defined as the factor by which the observed alpha half-life is greater than that calculated. Since even-even ground state alpha transitions are essentially unhindered, the hindrance factor for these transitions is defined equal to unity. Hindrance factors for transitions to excited states can be found in an article by Perlman and Rasmussen.[25] For odd-mass nuclei, alpha transitions to the ground state of the daughter are often highly hindered with an essentially unhindered transition leading to an excited state. The collective model considers these essentially unhindered excited state transitions to be those for which ΔI and ΔK are 0. On this basis, the 74.6-kev level in Np^{239} should be a 5/2- level; and the 117-kev and 173-kev levels, provided that they are members of the same rotational band, should be 7/2- and 9/2- levels respectively. By using the energies of the first three excited levels of Np^{239} in Eq. (4), one obtains a value $I_0 = 2.12$. It should be noted that this method of determining I_0 is extremely sensitive to small changes in the energy values of the excited levels.

According to the collective model, the transition probability for alpha decay from a parent in state I_i, K_i to a daughter state I_f, $K_f = K_i$ is given by

$$P = P_0(Z,E) \sum_L c_L \langle I_i L K_i O | I_i L I_F K_F \rangle^2 \quad , \tag{5}$$

where $P_0(Z,E)$ is an energy-dependence factor from alpha-decay theory, L is the angular momentum quantum number of the emitted alpha particle, c_L is the reciprocal of the hindrance factor for the alpha group of angular momentum L, and the brackets denote Clebsch-Gordan coefficients. Hummel[16] has used Eq. (5) to calculate intensity ratios of the first three excited levels in the decay of Am^{243} on the assumption that the spins of these levels are 5/2, 7/2, and 9/2. In Hummel's calculation the value of c_0 was taken as unity, since L = 0 transitions are assumed to be essentially unhindered. To determine the values of c_L to use with L = 2 or L = 4 transitions, an average of the observed L = 2 and L = 4 hindrance factors for neighboring even-even nuclei was taken. The intensity ratios calculated by Hummel are 100/12.0/1.9. These are in reasonable agreement with the experimentally observed ratios of 100/13.2/1.5.

For the transition from the 5/2- state in Am^{243} to the 5/2- state in Np^{239}, alpha multipolarites of 0, 2, and 4 are permitted by the selection rules for alpha decay. The value of the alpha-gamma angular correlation function is very sensitive to the mixing ratio* of these multipoles. It is

*The mixing ratio δ is defined such that the ratio of the total intensity of the L' pole to the intensity of the L pole is equal to δ^2.

commonly assumed that the contribution from L = 4 alpha waves is negligible. The intensity ratio of the L = 0 and L = 2 alpha waves can be determined from the equation

$$\frac{I_{L'}}{I_L} = \frac{C_{L'} < I_i L' K_i 0 | I_i L' I_f K_f >^2}{C_L < I_i L K_i 0 | I_i L I_f K_f >^2} \quad . \tag{6}$$

The value of c_0 is taken as unity, and a value of c_2 is obtained by taking an average of values of the L = 2 hindrance factors for the neighboring Pu^{242} and Cm^{244} nuclei. By using Eq. (6), an intensity ratio I_2/I_0 of 0.22 is obtained for alpha decay to the 5/2- level in Np^{239}. On the basis of this intensity ratio, and on the assumption that the gamma ray which de-excites the 5/2- level is E1, the theoretical alpha-gamma angular correlation function for the 5/2- → 5/2- → 5/2+ transition in the decay of Am^{243} to Np^{239} can be calculated from formulas given by Biedenharn and Rose.[6] For this transition, the theoretical correlation function is

$$W(\theta) = 1 - 0.36\ P_2(\cos\theta)$$

and the anisotropy value is A = -0.46.

B. Nuclear Properties of Even-even Nuclei

Decay-scheme diagrams for alpha decay of Th^{230}, U^{232}, and Ra^{226} are shown in Figs. 2, 3, and 4. The decay of Th^{230} to Ra^{226} has been investigated by Stephens et al.,[26,27] and by Hummel;[16] the decay of U^{232} to Th^{228} has been investigated by Asaro and Perlman,[28] and by Scharff-Goldhaber et al.;[29] the decay of Ra^{226} to Rn^{222} has been investigated by Asaro and Perlman,[30] and by Harbottle et al.[31] Table I gives half-life values for the first excited (2+) level in the alpha decay of Th^{230}, U^{232}, and Ra^{226}, together with data on the energies and conversion coefficients of the gamma radiation which depopulates this level. The half-lives have been measured by the delayed coincidence method by Bell et al.[32] For the 68-kev level in Ra^{226}, the half-life has been confirmed in a measurement by Vartapetian and Foucher.[33]

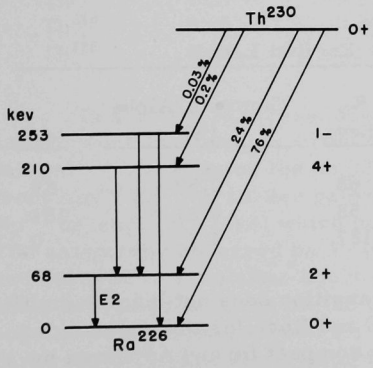

Fig. 2. Decay-scheme Diagram for the Alpha Decay of Th^{230} to Ra^{226}.

Nuclei which contain an even number of neutrons and an even number of protons have a ground state spin of zero.[34] In most instances (and, in particular, for nuclides used in this investigation) the first excited level of even-even nuclei is known to have spin 2, both on the basis of predictions from the collective model and as a result of measurements to

determine the multipolarity of the gamma radiation which de-excites this level. For even-even nuclei, the Bohr-Mottelson theory predicts a ground state spin of zero, with spins 2, 4, 6, etc., for the excited levels belonging to the same rotational band. By means of Eq. (4), the ratio of the energy of the second excited level to that of the first excited level for these nuclei is found to be 3.33. Perlman and Asaro[35] have tabulated this ratio for eleven even-even alpha emitters from Ra^{222} to Cm^{242} and have found excellent agreement with the prediction of the collective model. Based on measurements of their internal conversion coefficients, the 68-kev gamma in Ra^{226}, the 58-kev gamma in Th^{228}, and the 187-kev gamma in Rn^{222} are all found to be electric quadrupole.[36-38]

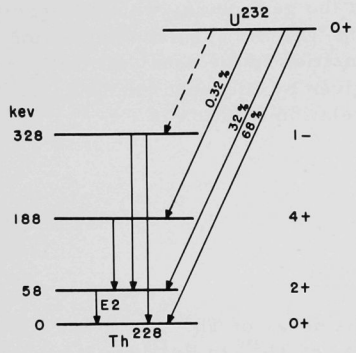

Fig. 3. Decay-scheme Diagram for the Alpha Decay of U^{232} to Th^{228}.

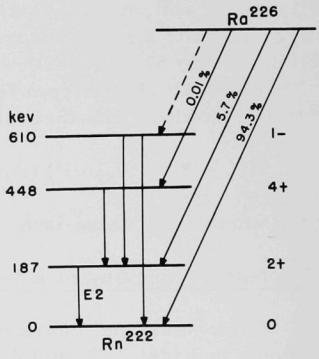

Fig. 4. Decay-scheme Diagram for the Alpha Decay of Ra^{226} to Rn^{222}.

Table I. Excited Level Half-lives, Gamma Energies, Gamma/Alpha Intensity Ratios, and Conversion Coefficients for the Gamma Transition from the 2+ Level Following Alpha Decay from Th^{230}, U^{232}, and Ra^{226}.

Parent Nuclide	Alpha-decay Half-life (years)	Excited Level: Half-life (sec x 10^{10})	Excited Level: E_γ (kev)	Excited Level: Gammas/Alpha (%)	Excited Level: e/γ
Th^{230}	8.0×10^4	6.3 ± 0.2	68	0.59	39
U^{232}	74	4.0 ± 0.3	58	0.21	151
Ra^{226}	1622	3.2 ± 0.2	187	4.7	0.22

Because the 0-2-0 alpha-gamma transition does not involve multipole mixtures, the equations of Biedenharn and Rose for the angular correlation function can be reduced to a more compact form. As shown by

Frauenfelder,[1] the coefficients A_n in the correlation function can be expressed as products of F coefficients. These F coefficients have been tabulated by Ferentz and Rosenzweig.[39] From the formulas of Frauenfelder together with the coefficients of Ferentz and Rosenzweig, the theoretical alpha-gamma angular correlation function for a 0-2-0 transition in which the alpha wave is pure L = 2 and the gamma multipolarity is also quadrupole is found to be

$$W(\theta) = 1 + 0.714\ P_2(\cos\theta) - 1.714\ P_4(\cos\theta) \quad .$$

C. Review of Experimental Results Reported in the Literature

The alpha-gamma angular correlation measurements reported in the literature have for the most part been carried out with solid sources on even-even nuclei. A summary of results obtained with even-even nuclei is presented in Table II. It is evident that, in general, the correlation is highly attenuated in solid sources.

Table II. Summary of Published Results for Alpha-Gamma Angular Correlation Measurements Made with Solid Sources on the 0-2-0 Transition in Even-even Nuclides. The theoretical correlation function is

$$1 + 0.714\ P_2(\cos\theta) - 1.714\ P_4(\cos\theta) \quad .$$

Parent Nuclide	A_2	A_4	G_2	G_4	Reference
Ra^{224}	0.534	-1.539	0.75	0.90	40
Ra^{226}	0.490	-1.299	0.69	0.76	40
Pu^{238}	0.330	-1.145	0.46	0.67	40
Th^{230}	0.338	-0.969	0.47	0.56	41
Th^{230}	0.27	-0.82	0.38	0.48	42
Th^{230}	0.25	-0.89	0.35	0.52	43
Th^{228}	0.30	-0.86	0.42	0.50	44

In addition to measurements with even-even nuclei, some solid source work has been reported for odd-A and odd-odd nuclei. Fraser and Milton[45] investigated the angular correlation between alpha particles from Am^{241} and the 60-kev gamma from Np^{237}. The 60-kev gamma from Np^{237} de-excites a level which has a relatively long half-life (6.3×10^{-8} sec). The anisotropy observed by Fraser and Milton had an initial value of -0.15 and decayed to zero after about 2×10^{-8} sec. The initial value is only about one-third as large as the theoretical value. Petit[46] measured the angular correlation between Pu^{239} alpha particles and the 51-kev gamma-ray from U^{235}. The level spins involved in this transition were presumed to be 1/2+, 5/2+, 1/2+, and the observed results could be accounted for by

assuming an attenuated correlation having attenuation coefficients $G_2 = 0.63$ and $G_4 = 0.49$. Horton[47] studied the angular correlation between Bi^{212} alpha particles and the 40-kev gamma ray from Tl^{208}. On the basis of the correlation which he observed, several different spin assignments are possible.

An investigation of perturbation of the alpha-gamma angular correlations in the decay of Am^{241}, Am^{243}, and Cm^{243} as a function of the material into which the daughter nucleus recoils has recently been reported by Flamm.[8] Flamm carried out experiments with thick sources of the active oxide, thin sources vaporized on both conducting and nonconducting media, and with samples in which the daughter nuclei were allowed to recoil into water and into vacuum. The results showed that the correlation is less attenuated when the daughter recoils take place in a conducting medium than when the recoils take place in a nonconducting medium. The correlation was found to be completely attenuated in an integral correlation experiment* on Am^{241} in which the daughter nuclei were allowed to recoil into distilled water.

In the only previously reported alpha-gamma angular correlation measurements involving liquid sources, Novey,[13] and Krohn, Novey, and Raboy[14] used delayed coincidence techniques to measure the angular correlation between alpha particles of Am^{241} and the 59.6-kev gamma ray which depopulates the 6.3×10^{-8}-sec level in Np^{237}. For HCl, $HClO_4$, and H_2SO_4 solutions in the range 1 to 3 molar, they found the attenuation coefficients to be independent of acid concentration and to decrease exponentially with time as predicted by theory. The observed relaxation constants were $\lambda_2 = 13.5 \pm 2$ per μsec for HCl and $HClO_4$, and $\lambda_2 = 21 \pm 2$ per μsec for H_2SO_4. By extrapolation of their data to zero time, Novey's group was able to obtain good agreement between measured and theoretical values of the anisotropy. Measurements were also made with liquid sources containing acetic acid, but these measurements did not give results which agreed with theory. Novey's group attributed the attenuation which was observed with acetic acid sources to the relatively greater complexing strength of acetate ions.

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*An integral correlation experiment is one in which the resolving time of the fast coincidence circuit is much longer than the half-life of the intermediate level. In such an experiment the "coincident" gamma rays come from nuclei which have experienced varying degrees of perturbation of the spin orientation of their intermediate levels due to the fact that they existed at the intermediate level for varying periods of time. The observed attenuation coefficient for an integral correlation experiment is the time average of $G_k(t)$. A differential or delayed coincidence measurement, on the other hand, is one for which the resolving time of the coincidence circuit is much shorter than the half-life of the intermediate nuclear level. To a first approximation, $G_k(t)$ is constant over the resolving time of the apparatus. In theory, by extrapolation of the data from a series of delayed coincidence measurements to zero time, it should be possible to obtain the true correlation.

D. Theory of Perturbation of the Angular Correlation

Attenuation of the angular correlation due to interactions between nuclear moments and extranuclear electric and magnetic fields has been treated theoretically by Goertzel,[48] Alder,[49] and Abragam and Pound.[50-52] An excellent summary of the theoretical work on attenuation of the angular correlation has been prepared by Steffen.[3] Perturbation of the angular correlation can result from interactions between the nuclear magnetic dipole moment and extranuclear magnetic fields and between the nuclear electric quadrupole moment and extranuclear electric field gradients. These interactions may be either static or time dependent.

A static interaction between a magnetic field H and the magnetic dipole moment of a nucleus, $\mu = gI\mu_N$, induces the nuclear angular momentum to precess at the Larmor frequency, $\nu_L = g\mu_N H/h$. If the direction of the magnetic field is different from the axis of quantization of the intermediate level angular momentum, and if the period of precession is sufficiently short, the magnetic sublevel population of the intermediate level will be altered. According to Kopfermann,[53] studies of optical hyperfine structure indicate that the magnetic field produced at the nucleus by the valence electrons may be as high as 10^5-10^6 oersteds. From measurements of hyperfine structure in the spectrum of americium, Manning, Fred, and Tomkins[54] have calculated a value of +1.4 nm for the nuclear magnetic dipole moment of Am^{243}. With this value of μ, the Larmor precession frequency of the Am^{243} nucleus in a field of 10^6 oersteds is found to be 0.43×10^9 cycles/second. Pake[55] tabulates ground state spins and nuclear magnetic moments in units of the nuclear magneton for 38 nuclides from H^1 to Tl^{205}. For these nuclides, the Larmor precession frequencies in a field of 10^6 oersteds would range from 0.1×10^9 to 5×10^9 cycles/second. A static interaction between magnetic fields and nuclear moments of the magnitudes indicated above could cause attenuation of the angular correlation function in nuclei with intermediate level lifetimes greater than a fraction of a millimicrosecond.

The interaction energy between the nuclear quadrupole moment Q and the field gradient of an electric field which has axial symmetry is given by

$$E_Q = \frac{3m_z^2 - I(I+1)}{4I(2I-1)} eQ \frac{\partial E_z}{\partial z} , \qquad (7)$$

where the direction of the axis of the nuclear quadrupole moment with respect to the field axis is specified by the magnetic quantum number m_z. For a static electric interaction, the characteristic precession frequencies can be expressed as integral multiples of fundamental precession frequencies, which are given by

$$\nu_e^0 = \frac{3}{4I(2I-1)} \frac{eQ}{h} \frac{\partial E_z}{\partial z} \quad \text{for integral I} \tag{8}$$

and

$$\nu_e^0 = \frac{3}{2I(2I-1)} \frac{eQ}{h} \frac{\partial E_z}{\partial z} \quad \text{for half-integral I.} \tag{9}$$

Values of quadrupole interaction frequencies $\left(\frac{eQ}{h} \frac{\partial E_z}{\partial z}\right)$ and nuclear quadrupole moments Q obtained from quadrupole resonance experiments with crystalline materials are given in Table III. The Q values are from a tabulation by K. F. Smith,[56] and the quadrupole interaction frequencies are from a tabulation by H. Kopfermann[57] [except for the data on In^{115}, which is from Simmons and Slichter[58]]. On the assumption that the electric fields present at the nuclei indicated in the table are axially symmetric, electric field gradients have been calculated. In general, the field gradients are of the order of 10^{18} v/cm^2.

Table III. Electric Field Gradients Deduced from Quadrupole Resonance Experiments on the Basis of an Axially Symmetric Electric Field.

Nucleus	Molecule	$\left\lvert \frac{eQ}{h} \frac{\partial E_z}{\partial z} \right\rvert$ (mc/sec)	Q (10^{-24} cm^2)	$\frac{\partial E_z}{\partial z}$ (v/cm^2)
In^{115}	In metal	45.19	0.834	2.24×10^{17}
Cu^{63}	Cu_2O	53.4	-0.157	1.41×10^{18}
As^{75}	As_4O_6	232.5	0.3	3.20×10^{18}
Sb^{121}	$SbCl_3$	489	-0.53	3.82×10^{18}
Hg^{201}	$HgCl_2$	724	0.45	6.65×10^{18}

Since large nuclear deformations are known to occur in nuclei with nucleon configurations far removed from closed shells (i.e., far removed from Z = 82, N = 126), it is to be expected that such nuclei will exhibit large quadrupole moments. The quadrupole interaction frequencies for such nuclei should therefore also be relatively large. For example, intrinsic quadrupole moments of 9.0 to 10.3 x 10^{-24} cm^2 have been reported from Coulomb excitation of U^{235}, Np^{237}, and Pu^{239};[59] these values correspond to spectroscopic quadrupole moments of 3.2 to 3.7 x 10^{-24} cm^2 for nuclei with I = 5/2. Kerman[60] gives a value of Q = 6 x 10^{-24} cm^2 for the quadrupole moment of U^{233}, and Manning, Fred, and Tomkins[54] give a value of Q = 4.9 x 10^{-24} cm^2 for the quadrupole moment of Am^{243}. The ground state spin for both U^{233} and Am^{243} is 5/2. Using a value of 10^{18} v/cm^2 for the electric field gradient, one obtains fundamental precession frequencies of 2.3 x 10^8 sec^{-1} for U^{233} and 1.9 x 10^8 sec^{-1} for Am^{243}. At such interaction frequencies, electric

interactions are also capable of perturbing the angular correlation in nuclides for which the intermediate level lifetime is greater than a fraction of a millimicrosecond.

In a polycrystalline solid, the attenuation coefficients G_k for a static interaction never go to zero. This is a consequence of the fact that in a polycrystalline material there are always some nuclei for which the axis of spin quantization and the direction of the precession-inducing electric or magnetic field coincide. For these nuclei, precession of the spin vector does not induce changes in the magnetic sublevel population. It has been shown by Alder[49] and by Abragam and Pound[52] that the minimum or "hard core" value of G_k for an integral correlation which is perturbed by an axially symmetric static interaction is $G_k = 1/(2k+1)$.

Expressions for the attenuation coefficients as a function of precession frequency and time have been derived by Alder[49] for static magnetic interactions and by Abragam and Pound[52] for static electric interactions involving axially symmetric fields. For a magnetic interaction, G_2 is always greater than G_4. However, for an electric interaction, G_4 is greater than G_2 for certain values of intermediate-level angular momentum. This is the case for the 0-2-0 transition in even-even nuclei (for which the intermediate level has spin I = 2). The experimental observation that G_4 is greater than G_2 in the perturbed alpha-gamma angular correlation function obtained with solid sources for even-even nuclei indicates that for these nuclei the predominant factor in the attenuation of the correlation is an interaction between the electric quadrupole moment of the nucleus and extranuclear electric field gradients.

Time-dependent interactions involving electric or magnetic fields which fluctuate randomly in direction (as in a liquid) induce changes in magnetic sublevel populations defined with respect to any direction of the axis of quantization. Hence, in contrast to static interactions, time-dependent interactions can completely wipe out the angular correlation if the lifetime of the intermediate nuclear level is sufficiently long. The approach to an isotropic condition is exponential, with the attenuation coefficients being given by $G_k(t) = e^{-\lambda_k t}$. For a given liquid, λ_k is proportional to the correlation time τ_c of the perturbing field in the liquid. If the correlation time is very short so that λ_k is small, G_k will decrease slowly with time. Thus, for an appropriate liquid and for decays involving short, excited level lifetimes, the possibility exists of observing a correlation which is not significantly attenuated.

In an integral angular correlation measurement where the lifetime τ_N of the intermediate nuclear level is much shorter than the resolving time of the coincidence apparatus, the experimentally observed attenuation coefficient for a time-dependent interaction is given by

$$G_k = \frac{1}{\tau_N} \int_0^\infty e^{-t/\tau_N} \, G_k(t)dt = \frac{1}{1 + \lambda_k \tau_N} \quad . \tag{10}$$

Abragam and Pound[52] have shown that for a time-dependent quadrupole interaction, λ_k is given by

$$\lambda_k = \frac{3}{80} \left(\frac{eQ}{\hbar}\right)^2 \left\langle \left(\frac{\partial E_z}{\partial z}\right)^2 \right\rangle_{av} \tau_c \, \frac{k(k+1)[4I_B(I_B+1) - k(k+1) - 1]}{I_B^2 (2I_B - 1)^2} \quad , \tag{11}$$

where

$$\left(\frac{eQ}{\hbar}\right)^2 \left\langle \left(\frac{\partial E_z}{\partial z}\right)^2 \right\rangle_{av}$$

is the mean-square value of the quadrupole interaction and I_B is the intermediate-level spin.

For a time-dependent magnetic interaction caused by a continual random change in the orientation of the atomic electron spin for a paramagnetic ion, Abragam and Pound have shown that λ_k is given by

$$\lambda_k = \frac{2}{3} \tau_s \omega_s^2 I_B(I_B+1) S(S+1) \left[1 - (2I_B+1) W(I_B 1 k I_B \,|\, I_B I_B)\right] \quad , \tag{12}$$

where τ_s is the relaxation time of the electron spin, ω_s is the interaction frequency between the nucleus and the magnetic moment due to the electron spin, and W is a Racah coefficient.

III. EXPERIMENTAL PROCEDURE

A. Preparation of Sources

The sources employed in these alpha-gamma angular correlation measurements consisted of solid Am^{243} sources and liquid sources containing either Am^{243}, Th^{230}, U^{232}, or Ra^{226} in dilute perchloric acid solutions.

Solid Am^{243} sources were prepared by vaporizing the active material on suitable backing material which was cemented to an aluminum ring. A summary of information on source thickness and backing material for the solid sources is given in Table IV. Presumably the americium in the sources was in the form Am_2O_3. The diameter of the active area of each of the solid sources was $\frac{3}{8}$ in. The sources were covered with aluminum or gold foils to catch the neptunium recoils and prevent radioactive contamination of the counting chamber. All solid source experiments were conducted in vacuum.

Table IV. Summary of Data on The Preparation of Solid Am^{243} Sources

		Backing Material		Cover	
Source No.	Source Thickness ($\mu g/cm^2$)	Type	Thickness (mg/cm^2)	Type	Thickness ($\mu g/cm^2$)
1	46.8	aluminum	6.86	aluminum	200
3	5.7	aluminum	6.86	aluminum	200
4	3.9	gold	0.40	aluminum	200
5	5.8	mica	2.8	aluminum	200
6	3.2	gold	2.68	gold	150

Liquid sources were prepared by placing a drop containing 2 or 3 μliters of solution on a rubber hydrochloride membrane cemented to an aluminum ring. A microscope cover glass, 1 cm^2 in circular cross section, was then placed over the drop. The thickness of the rubber hydrochloride membranes was such that their stopping power for alpha particles was equivalent to 0.8-1.0 mg/cm^2 of aluminum. During the time that data were being obtained, the sources were mounted vertically in a sealed aluminum tank. In order to prevent a liquid source from slipping downward, a line of high-vacuum grease was painted across the rubber hydrochloride membrane directly beneath the cover glass. In order to keep a source in the liquid state during an experiment, the floor of the aluminum counting chamber was covered to a depth of $\frac{1}{4}$ in. with a solution of water and glycerol, and the wall of the chamber was lined with blotting paper which dipped into the water-glycerol solution. The glycerol content of the chamber solution was adjusted to give the solution approximately the same vapor pressure as that of the liquid source.

All liquid source experiments were carried out in a helium atmosphere. After a source had been placed in the counting chamber, the chamber was flushed at the rate of one chamber volume per 5 min with helium which had been passed through two gas bubblers containing heated distilled water. The helium flush was continued for 16 min and the chamber was then sealed. With this procedure, it was possible to keep a source in the liquid state for as long as two weeks.

Details of procedures employed in the chemical purification of the radionuclides used in these experiments and data on the purity of the solutions from which sources were prepared are given in Appendix II.

B. Counting Chamber and Detectors

Figure 5 is a diagram of the counting chamber showing the positions of the radioactive sample and of the alpha and gamma detectors. The gamma detector located outside the counting chamber was fixed in position. The alpha detector located inside the counting chamber was optically coupled to its photomultiplier tube through a glass window in the side of the chamber. The radioactive sample was supported on a stand at an angle of 45° with respect to the alpha detector. Changing of the angle θ between alpha and gamma detectors was accomplished by rotating the counting chamber about a vertical axis through the center of the chamber. By means of adjustment screws in the base of the sample support stand it was possible to center the sample on the axis of rotation of the counting chamber.

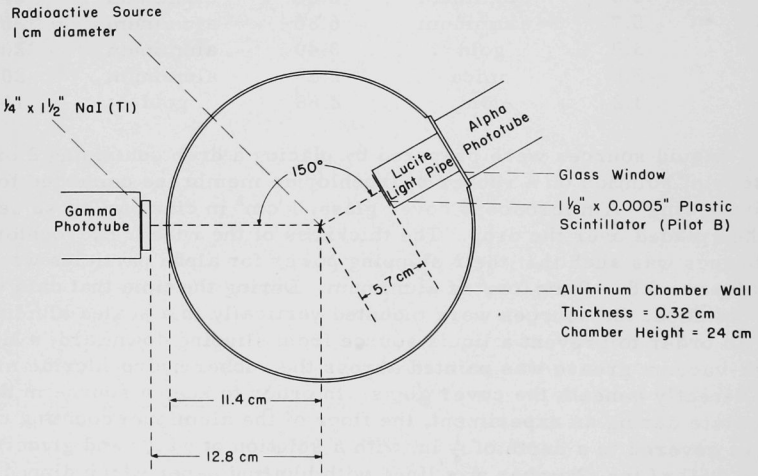

Fig. 5. Diagram of Counting Chamber Showing Positions of Alpha and Gamma Detectors. The alpha detector shown is that used for liquid sources.

For measurements with Am^{243} and Th^{230}, the gamma detector was a $\frac{1}{4}$-in.-thick by $1\frac{1}{2}$-in.-diameter NaI(Tl) crystal coupled to an RCA-6342 photomultiplier tube. For measurements with U^{232} a $\frac{3}{8}$-in.-thick by 2-in.-diameter crystal was used. Because of the higher gamma energy involved, a 1-in.-thick by $1\frac{1}{2}$-in.-diameter crystal was used for the Ra^{226} measurements.

A $\frac{3}{8}$-in.-thick by $1\frac{1}{4}$-in.-diameter anthracene crystal was employed as alpha detector for the solid source experiments. The Np^{239} daughter from alpha decay of Am^{243} is a beta emitter, with a 2.3-day half-life, which quickly grows into secular equilibrium with Am^{243}. As seen in Fig. 6, when solid americium sources were used and the counting chamber was evacuated, the anthracene crystal could easily resolve the alpha peak from the beta background. However, in the liquid source measurements, the alpha particles were degraded in energy in traversing the liquid source, the rubber hydrochloride membrane, and the approximately 6 cm of helium between source and detector. In the thick anthracene crystal, neptunium betas would have given the same size pulses as those produced by the degraded alphas. Therefore, for liquid source studies, the anthracene crystal was replaced by a 0.5-mil-thick plastic scintillator (Pilot B). Figure 7 shows the Am^{243} alpha spectrum obtained for a liquid source with the plastic scintillator. The dashed curve in Fig. 7 indicates the neptunium beta spectrum obtained with the same sample by admitting air into the counting chamber.

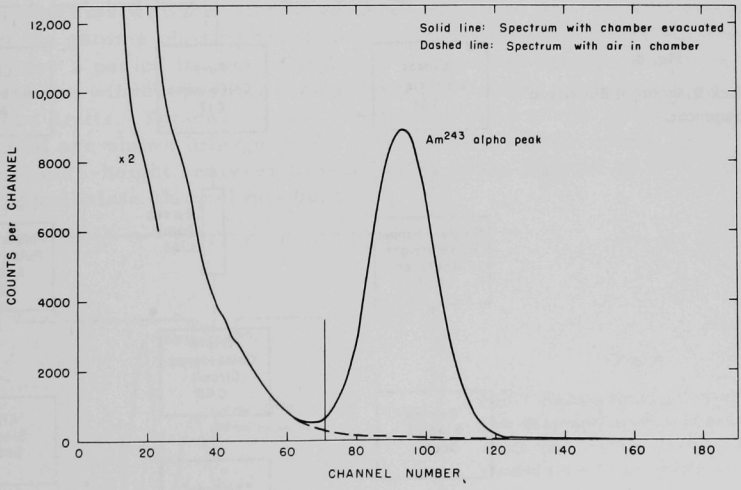

Fig. 6. Am^{243} Alpha Spectrum (solid source). The vertical line at channel number 72 indicates the integral bias setting on the single channel pulse height analyzer.

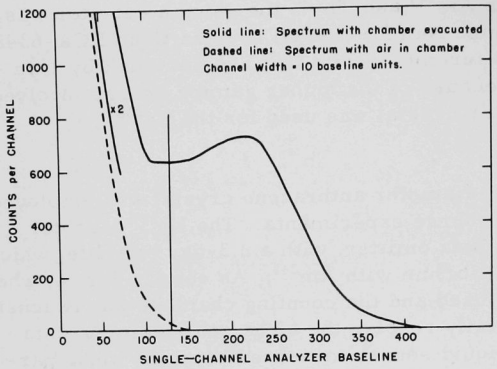

Fig. 7.

Am^{243} Alpha Spectrum (liquid source). The vertical line at baseline 130 indicates the integral bias setting on the single-channel pulse-height analyzer.

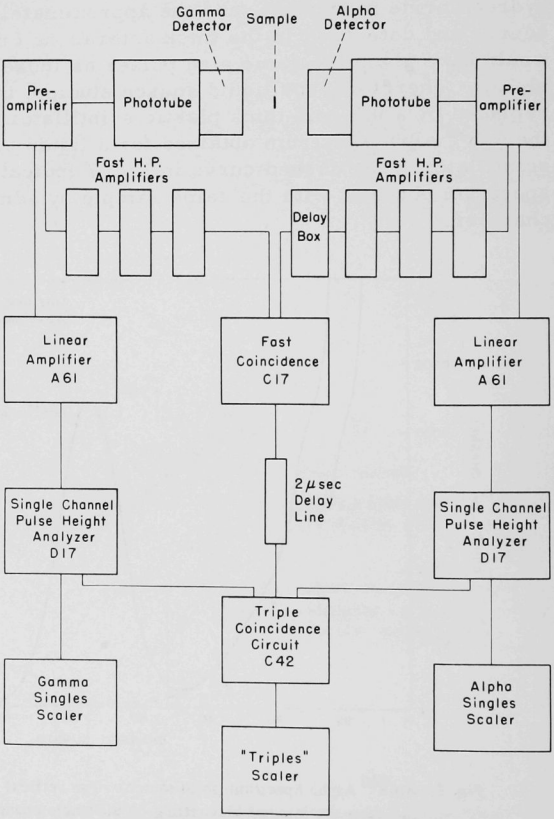

Fig. 8.

Block Diagram of Electronic Equipment.

C. Electronic Equipment

A block diagram of the electronic equipment used for these measurements is shown in Fig. 8. The resolving time τ of the fast coincidence circuit was 17 mμsec.

To obtain the angular correlation function for a given sample it is necessary to know the prompt alpha-gamma coincidence rate as a function of the angle between the alpha detector and the gamma detector. However, not all prompt coincidence events are the result of coincidences between alpha particles and gamma photons belonging to the particular transition being investigated. In order to insure that only the desired prompt coincidence events were counted, a triple coincidence was required between prompt coincidence pulses and alpha and gamma singles pulses of the appropriate pulse heights.

The alpha single-channel pulse-height analyzer was used on integral bias with the bias setting adjusted to discriminate against beta pulses and to reject noise pulses which only serve to increase the chance coincidence rate. Typical alpha spectra for both solid and liquid sources are shown in Figs. 6 and 7. The bias setting on the alpha single-channel pulse-height analyzer is indicated in the figures by a vertical line at the appropriate channel number.

The gamma single-channel pulse-height analyzer was operated on differential bias with the window adjusted to accept only the full energy peak of the gamma photons being investigated. Whenever a sample was counted for a period in excess of 24 hr, the gamma spectrum was checked daily and the window position was adjusted to compensate for amplifier and phototube drifts. Typical gamma spectra for each of the radioisotopes investigated are shown in Figs. 9-12. The window widths on the single-channel pulse-height analyzer are indicated in the figures by vertical lines at the appropriate channel numbers.

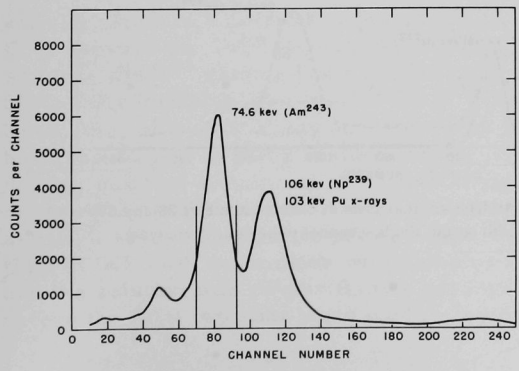

Fig. 9.

Am^{243} Gamma Spectrum. The vertical lines at channel numbers 70 and 92 indicate the window width on the single-channel pulse-height analyzer.

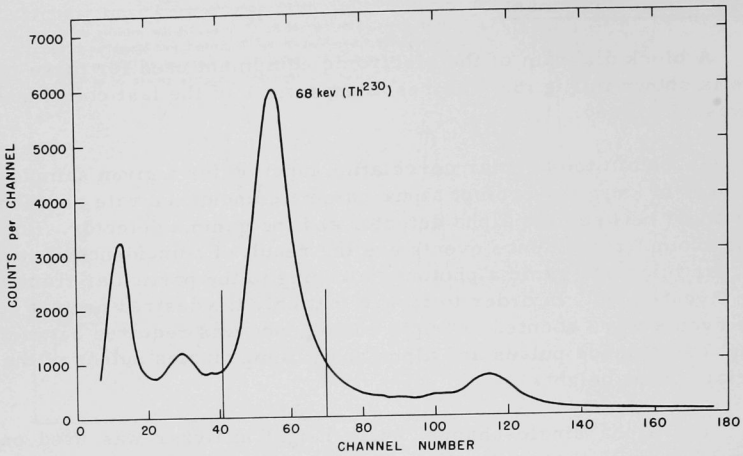

Fig. 10. Th^{230} Gamma Spectrum. The vertical lines at channel numbers 41 and 70 indicate the window width on the single-channel pulse-height analyzer.

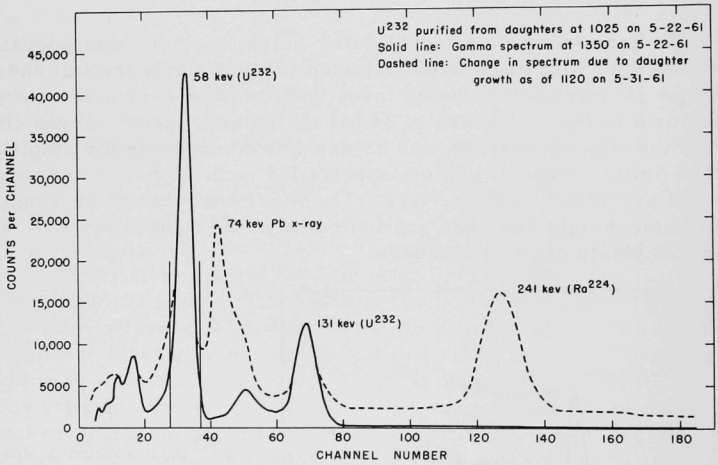

Fig. 11. U^{232} Gamma Spectrum. The vertical lines at channel numbers 28 and 37 indicate the window width on the single-channel pulse-height analyzer.

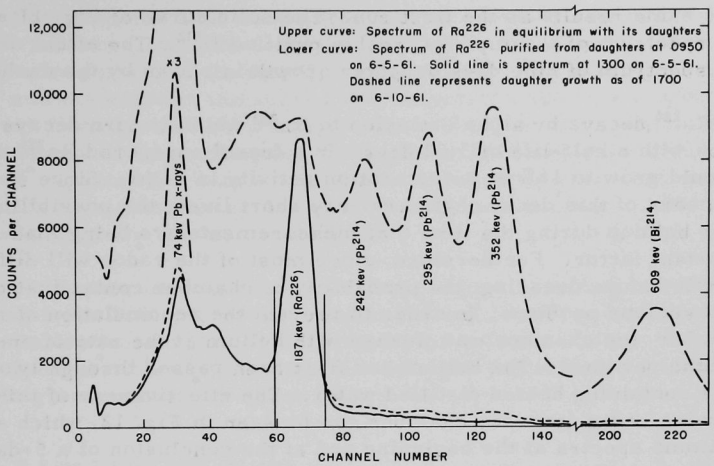

Fig. 12. Ra^{226} Gamma Spectrum. The vertical lines at channel numbers 59.5 and 74 indicate the window width on the single-channel pulse-height analyzer.

Am^{243} decays by alpha emission to Np^{239} which then decays by beta emission, with a 2.3-day half-life, to Pu^{239}. In the parent solution from which the americium samples were made, Np^{239} grew into secular equilibrium in a few days. However, because Pu^{239} has a 24,000-year half-life, the growth of other daughters into this solution was negligible. The Ra^{226} daughter from alpha decay of Th^{230} has a half-life of 1622 years. Hence, it was not necessary to be concerned about daughter growth in the thorium solution after the thorium was once purified from its daughter products.

U^{232} decays by alpha emission to Th^{228} which in turn decays by alpha emission with a half-life of 1.91 years. In a freshly prepared U^{232} sample, Th^{228} will grow to one percent of its saturation activity after 10 days. However, after 10 days, the 84-kev gamma from the decay of Th^{228} will have an activity which is 8 percent of that of the 58-kev gamma from the decay of U^{232}, because the Th^{228} gamma has a gamma-alpha intensity ratio of 1.6% whereas the U^{232} gamma has a gamma/alpha intensity ratio of only 0.2%. Since the remaining members of the U^{232} decay chain are all very short lived, the ratio of K X-ray activity to the 58-kev gamma activity soon becomes large. In just a matter of days, the growth of daughters into a freshly purified solution of U^{232} could affect the measured correlation because of the possibility of observing coincidences between alpha particles and K X rays. In order to prevent this, only two runs occupying a total time of one week were made with samples from a freshly purified solution, and the solution was re-purified by passage through an ion exchange column before the final two runs were made. In each set of runs, the second run

gave the same results as the first run. The solid curve of Fig. 11 shows the gamma spectrum of a sample of freshly purified U^{232}. The effect on the gamma spectrum of nine days daughter growth is shown by the dashed line.

Ra^{226} decays by alpha emission to Rn^{222} which in turn decays by alpha emission with a half-life of 3.82 days. In a freshly prepared Ra^{226} sample, Rn^{222} would grow to 16% of its saturation activity in 24 hr. Since succeeding members of this decay chain are very short lived, the possibility of daughter buildup during the time that measurements are being made becomes an important factor. Furthermore, since most of the radon will diffuse from the sample before decaying, the probability of chamber contamination becomes a serious problem. In order to prevent the accumulation of radon in the chamber, the chamber was flushed with helium at the rate of one chamber volume per hour. The helium had first been passed through two gas bubblers containing heated distilled water. The effectiveness of this method of removing radon from the chamber can be seen in Fig. 12, which shows Ra^{226} gamma spectra at the beginning and at the conclusion of a 5-day measurement. The gamma spectrum of Ra^{226} in secular equilibrium with its daughters is shown in the same figure for purposes of comparison.

Because of the symmetry of the angular correlation function, it is sufficient to make measurements in the quadrant between $\theta = 90°$ and $\theta = 180°$. The accumulation and the recording of data were done automatically by means of a turntable mechanism which allowed the angles at which counts were to be made and the time of the counts to be preselected. For most of the runs (except for several runs with liquid sources containing Am^{243}, for which data were obtained only at 90° and at 180°) data were recorded at angular intervals of 15° over the range from 90° to 180° inclusive. At the end of a counting interval, data consisting of alpha singles, gamma singles, and triple-coincidence counts were printed automatically on adding machine tapes and the turntable advanced automatically to the next angle. At the completion of a cycle, the turntable reversed itself and repeated the cycle in the opposite sense.

D. Treatment of Data

Several corrections were made to the raw triple-coincidence data before using them to compute the angular correlation. First, the triples data were corrected for chance coincidences and for background coincidences (beta-gamma coincidences in the case of the Am^{243} measurements, and coincidences resulting from chamber contamination which was present during a part of the Th^{230} measurements). At the high counting rates observed with the Am^{243} samples, coincidences due to cosmic ray showers were not significant. At the lower counting rates obtained with even-even alpha emitters, cosmic ray events were significant. For the experiments with even-even alpha emitters, the equipment was covered with a cosmic ray shield consisting of an umbrella of Geiger tubes, which was operated in anti-coincidence with the triple-coincidence circuit.

Secondly, the data were corrected for coincidences resulting from gamma radiation scattered from the walls of the aluminum counting chamber. The magnitude of the scattering correction was determined by interposing midway between the source and the gamma detector a lead disk of sufficient diameter and thickness to absorb the gamma radiation from the direct beam and then measuring the triple-coincidence rate. This procedure was repeated for each angle at which measurements were made. The correction for coincidences due to scattered radiation was found to be about 5-10% of the gross triples counting rate.

No correction was made for small-angle scattering of alpha particles resulting from Coulomb interactions in the liquid source and the rubber hydrochloride membrane. According to Rutherford's theory of the scattering of alpha particles,[61] the fraction of alpha particles scattered through an angle greater than ϕ is given by

$$q = \frac{1}{4} \pi \, ntb^2 \cot^2 \frac{\phi}{2} \quad ,$$

where

$$b = 4Ze^2/MV^2 \quad .$$

Here, n is the number of atoms per unit volume of the scattering medium and t is the thickness of the scattering medium; Z is the atomic number of the scattering medium, and M and V refer to the mass and velocity of the incident alpha particle. Calculations were made to determine the amount of scattering experienced by 5-Mev alpha particles in passing through 2 mg/cm^2 of water plus 1 mg/cm^2 of aluminum (i.e., through 2 λ of solution plus a rubber hydrochloride membrane with stopping power equivalent to 1 mg/cm^2 of aluminum). The calculations showed that only 0.3% of the alpha particles would be deflected more than 10°, 1.2% would be deflected more than 5°, and 7.6% would be deflected more than 2°. These values represent maximum scattering since in making the calculations it was assumed that all of the alpha particles traverse the 2 λ of solution. Since the half angle subtended at the source by the alpha detector is 14°, the effect of the scattering of alpha particles in the source and in the rubber hydrochloride membrane should be negligible.

Finally, the triples data were corrected for variations in alpha and gamma singles counting rates. This correction (1) served as a first-order correction for the fact that the source was sometimes not perfectly centered, and (2) corrected for amplifier drift in the singles circuits.

In order to obtain the angular correlation coefficients from the corrected triples data, the americium data were fitted to a function of the form $B_0 + B_2P_2(\cos\theta)$, and the even-even nuclide data were fitted to a function of

the form $B_0 + B_2P_2(\cos\theta) + B_4P_4(\cos\theta)$ by the method of least squares. The coefficients obtained by means of the least-squares computations were then corrected for the finite solid angles subtended at the source by the alpha and gamma detectors.

Rose[62] has shown that if the true angular correlation of a given radiation with respect to a fixed axis is given by

$$W(\theta) = 1 + \sum_n A_nP_n(\cos\theta); \; n = 2,4,\ldots,$$

then the measured correlation will be of the form

$$W(\theta) = B_0 + \sum_n B_nP_n(\cos\theta); \; n = 2,4,\ldots,$$

where

$$\frac{B_n}{B_0} = \frac{J_n}{J_0}A_n \quad .$$

Thus, for a correlation involving two radiations, the true coefficients can be obtained from the measured coefficients by means of the equation

$$A_n = \frac{B_n/B_0}{[J_n(1)/J_0(1)][J_n(2)/J_0(2)]} \quad , \tag{13}$$

where $J_n(1)$ and $J_n(2)$ refer to the first and second detectors, respectively. Expressions for J_n/J_0 for the case of gamma radiation have been derived by Rose. Numerical results based on Rose's equations and obtained with the aid of electronic computers have been published, for a number of different detector geometries, by Stanford and Rivers[63] and by West.[64]

The geometry factors used to correct these data for the finite size of the gamma detectors were obtained by interpolation of the numerical results of Stanford and Rivers. To correct the data for the finite size of the alpha detectors, the assumption was made that, since all alpha particles incident on the face of the alpha crystal produce pulses, the correction to be used for a given geometry should be equal to the correction for low-energy gamma radiation at that geometry. Correction factors are tabulated in Table V, in which $J_n(1)$ refers to the gamma detector and $J_n(2)$ to the alpha detector.

Table V. Correction Factors Used to Correct the Measured Angular Correlation Coefficients for the Finite Size of the Alpha and Gamma Detectors

Nuclide	$J_2(1)/J_0(1)$	$J_4(1)/J_0(1)$	$J_2(2)/J_0(2)$	$J_4(2)/J_0(2)$
Am^{243} (solid source)	0.98		0.95	
Am^{243} (liquid source)	0.98		0.96	
Th^{230}	0.98	0.94	0.96	0.86
U^{232}	0.97	0.91	0.96	0.86
Ra^{226}	0.98	0.95	0.96	0.86

IV. RESULTS

A. Am^{243} - Solid Sources

The alpha-gamma angular correlation between alpha particles from Am^{243} and the 74.6-kev gamma from Np^{239} has been studied both for a thick solid source and for thin sources vaporized on conducting and nonconducting backing materials. Data on the preparation of the solid sources is presented in Table IV of Chapter III. Since, as shown in Appendix I, the mean range of a neptunium recoil atom in Am_2O_3 is approximately 11.8 $\mu g/cm^2$, most of the recoils in source No. 1 (the thick source) were stopped in the americium oxide. The maximum path lengths traversed by recoil atoms in the thin sources ranged from 4.5 to 8.2 $\mu g/cm^2$. Therefore, most of the recoil daughters in these sources escaped from the active oxide into the backing material.

In obtaining data for the solid sources, the angle between the alpha detector and the gamma detector was varied in 15° steps over the interval from 90° to 180° inclusive. The angular settings were accurate to ± 0.5°. The counting time was made the same at each angle and was adjusted to give for each measurement a standard deviation in the coincidence counting rate of 2-4 percent. For some of the sources the counting time at each angle was 20 min and for other sources it was 30 min. For a complete run, the counting cycle was repeated from five to seven times so as to obtain a standard deviation of approximately one percent in the total coincidence counting rate at each angle.

Representative data from an experiment in which the neptunium recoils went into a conducting medium (aluminum) and from one in which the recoils went into a nonconducting medium (mica) are presented in Tables VI and VII. In these tables, complete data for a typical 30-min count at each angle are presented. The scaling factors shown are those actually used in obtaining the data. For the coincidence data, totals for the entire run are also given. The fractional standard deviations (FSD) for the triple-coincidence data were determined by means of the equations

$$\mathrm{FSD} = \sigma/\bar{x}_{\mathrm{net}} \quad , \tag{14}$$

where

$$\sigma = \sqrt{\sigma_1^2 + \sigma_2^2} \quad ; \tag{14a}$$

$$\sigma_1 = \sqrt{\frac{1}{n(n-1)} \sum_1^n (x_i - \bar{x})^2} \quad ; \tag{14b}$$

$$\sigma_2 = \sqrt{y_i/n} \quad . \tag{14c}$$

Here the subscript (1) refers to the gross triple-coincidence data and the subscript (2) refers to the spurious triples data. A single gross triples count is designated x_i, the correction for spurious coincidences applied to a single triples count is designated y_i, and n is the number of measurements in the run.

Table VI. Representative Data from an Angular Correlation Experiment with a Thin Solid Am^{243} Source on Aluminum Backing

Run No. 12
Activity: Am^{243}; 5.71 $\mu g/cm^2$ of Am_2O_3
Backing Material: aluminum; thickness = 0.001 in.

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	30	30	30	30	30	30	30
Gross alpha singles ($x10^{-3}$)	710	707	707	705	707	706	707
Alpha background ($x10^{-3}$)	6	6	6	6	6	6	6
Net alpha singles ($x10^{-3}$)	703	701	701	699	701	700	701
Gross gamma singles ($x10^{-2}$)	1434	1430	1425	1430	1428	1431	1441
Gamma background ($x10^{-2}$)	53	53	53	53	53	53	53
Net gamma singles ($x10^{-2}$)	1381	1377	1372	1377	1375	1378	1378
Gross triple coincidences ($x10^{-1}$)	185	183	178	157	150	134	135
Chance and beta-gamma coincidence ($x10^{-1}$)	1	1	1	1	1	1	1
Coincidence due to scattered radiation ($x10^{-1}$)	11.4	11.3	11.1	10.7	10.5	10.3	10.2
Net triple coincidences ($x10^{-1}$)	172.6	170.7	165.9	145.3	138.5	122.7	123.8
TOTALS							
Number of measurements	6	6	6	6	6	6	6
Total counting time (min)	180	180	180	180	180	180	180
Gross triple coincidences	11050	10870	10230	9550	8920	8350	8110
Spurious triple coincidences	742	739	726	703	690	678	670
Net triple coincidences	10308	10131	9504	8847	8230	7672	7440
Standard deviation (%)	1.6	0.5	1.6	1.0	0.6	1.8	1.1

Table VII. Representative Data from an Angular Correlation Experiment with a Thin Solid Am^{243} Source on Mica Backing

Run No. 21
Activity: Am^{243}; 5.82 $\mu g/cm^2$ of Am_2O_3
Backing Material: mica; thickness = 2.8 mg/cm^2

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	30	30	30	30	30	30	30
Gross alpha singles ($x10^{-3}$)	756	756	756	757	755	758	755
Alpha background ($x10^{-3}$)	6	6	6	6	6	6	6
Net alpha singles ($x10^{-3}$)	750	750	750	751	749	752	749
Gross gamma singles ($x10^{-2}$)	1455	1460	1460	1472	1469	1470	1469
Gamma background ($x10^{-2}$)	61	61	61	61	61	61	61
Net gamma singles ($x10^{-2}$)	1394	1399	1399	1411	1408	1409	1408
Gross triple coincidences ($x10^{-1}$)	174	177	164	161	159	159	162
Chance and beta-gamma coincidence ($x10^{-1}$)	1	1	1	1	1	1	1
Net triple coincidences ($x10^{-1}$)	173	176	163	160	158	158	161
TOTALS							
Number of measurements	6	6	6	6	6	6	6
Total counting time (min)	180	180	180	180	180	180	180
Gross triple coincidences	11090	10900	10900	10320	10320	10360	10270
Spurious triple coincidences	60	60	60	60	60	60	60
Net triple coincidences	11030	10840	10840	10260	10260	10300	10210
Standard deviation (%)	1.2	0.8	1.4	1.0	1.0	1.3	0.5

After correcting the triple-coincidence data for spurious coincidences and adjusting for variations in the alpha and gamma singles counting rates, a least-squares fit to the function $B_0 + B_2P_2 (\cos\theta)$ was made. The least-squares computation was made with the aid of an IBM-650 computer program, and the results were checked by manual calculations. In making the least-squares computation, data for each angle were given equal weight. The angular correlation functions obtained from the data presented in Tables VI and VII are shown in Figs. 13 and 14. In these figures, corrections for detector solid angle and the correction for the effect of alpha transitions to the 117-kev level in Np^{239} have not been made.

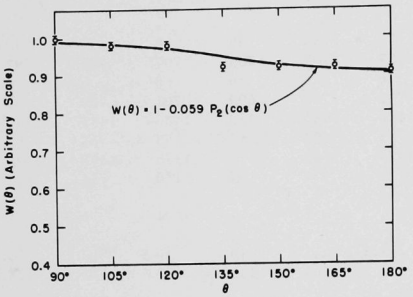

Fig. 13. Experimental Anisotropy for Thin Solid Am^{243} Source on Mica Backing. The curve was obtained by making a least-squares fit of the data to the function $W(\theta) = 1 + A_2P_2(\cos\theta)$. The correction for detector geometry has not been made.

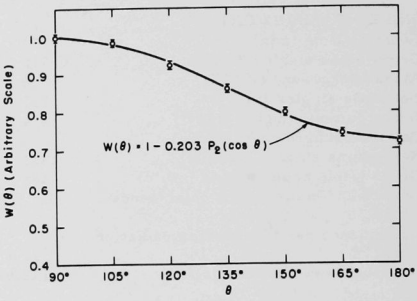

Fig. 14. Experimental Anisotropy for Thin Solid Am^{243} Source on Aluminum Backing. The curve was obtained by making a least-squares fit of the data to the function $W(\theta) = 1 + A_2P_2(\cos\theta)$. The correction for detector geometry has not been made.

Alpha decay to the 117-kev level in Np^{239} followed by highly converted M1 or E2 gamma radiation leading from the 7/2- to the 5/2- level accounts for 11.7% of the population of the 74.6-kev level in Np^{239}. The alpha-gamma angular correlation observed experimentally with all alphas and 74.6-kev gamma photons is thus a weighted average of the correlation involving the $5/2 \rightarrow 5/2 \rightarrow 5/2$ double cascade and the $5/2 \rightarrow 7/2 \rightarrow 5/2 \rightarrow 5/2$ triple cascade with intermediate radiation unobserved. Krohn et al.[14] have calculated the anisotropy for the triple cascade on the assumption that the alpha particle carries off two units of angular momentum and that the transition between the 7/2- state and the 5/2- state is either dipole or quadrupole. The calculated anistropries are -0.008 and +0.004, respectively, i.e., essentially isotropic. Flamm[8] reports a value for the anisotropy of the triple cascade of +0.06 based on pure L = 2 alpha waves and an E2/M1 mixing ratio of 1.7 for the unobserved radiation. If the triple cascade is assumed to be isotropic, the correction factor by which the experimental values of A_2 must be multiplied is 1.13 for Am^{243}.

Results for all of the Am^{243} solid source measurements are given in Table VIII. The values of A_2 have been corrected for the finite size of the alpha and gamma detectors and for the isotropic contribution of the triple cascade. The errors shown represent the standard deviations of the coefficients as determined by the IBM-650 least-squares program. Values for the anisotropy A have been derived from the experimentally determined results for A_2. The values for the attenuation coefficient G_2 have been calculated on the basis that the unattenuated value for A_2 is -0.36. The indicated errors in the average values of G_2 are only statistical and do not take into account any systematic errors (i.e., errors due to angular settings of the equipment or to the finite size of the source and the detectors).

Table VIII. Results for Am^{243} Solid Sources

Experiment Number	Source Number	Source Thickness ($\mu g/cm^2$ of Am_2O_3)	Recoil Stopping Material	A_2	A	G_2
6	1	46.8	Am_2O_3	-0.102 ± 0.003	-0.15	0.28
7	1	46.8	Am_2O_3	-0.104 ± 0.003	-0.15	0.29
8	1	46.8	Am_2O_3	-0.103 ± 0.002	-0.15	0.29
						ave = 0.29 ± 0.01
12	3	5.7	aluminum	-0.245 ± 0.005	-0.33	0.68
17	3	5.7	aluminum	-0.241 ± 0.011	-0.32	0.67
19	3	5.7	aluminum	-0.231 ± 0.006	-0.31	0.64
23	3	5.7	aluminum	-0.244 ± 0.007	-0.33	0.68
						ave = 0.67 ± 0.01
13	4	3.9	gold	-0.202 ± 0.004	-0.28	0.56
15	4	3.9	gold	-0.184 ± 0.014	-0.25	0.51
16	4	3.9	gold	-0.178 ± 0.011	-0.24	0.49
18	4	3.9	gold	-0.188 ± 0.006	-0.26	0.52
22	4	3.9	gold	-0.211 ± 0.008	-0.29	0.59
						ave = 0.54 ± 0.02
14	5	5.8	mica	-0.069 ± 0.006	-0.10	0.19
20	5	5.8	mica	-0.073 ± 0.004	-0.11	0.20
21	5	5.8	mica	-0.072 ± 0.012	-0.10	0.20
						ave = 0.20 ± 0.02
24	6	3.2	gold	-0.170 ± 0.012	-0.24	0.47
26	6	3.2	gold	-0.183 ± 0.008	-0.25	0.51
						ave = 0.49 ± 0.02

B. Am^{243} - Liquid Sources

Angular correlation measurements were made with liquid sources containing Am^{243} in dilute perchloric acid solutions for which the acid concentrations were 0.53M, 1.0M, and 3.0M. Data for some of the sources were obtained by following the same procedure as was used in the solid source experiments. However, because of the smaller activity of the liquid sources, it was necessary to count them for a longer time interval (one hour) at each

angle and to increase the number of cycles in order to achieve the same statistical accuracy as had been obtained with the solid sources. Because of the time requirement, most of the liquid-source data for Am^{243} were obtained by counting only at 90° and 180°. The anisotropy was then computed from the corrected data by means of Eq. (2). In runs for which counting was done only at 90° and 180°, the minimum number of measurements made at each angle was ten. Computations made for runs from which data were obtained at 15° intervals showed that the anisotropy determined from a least-squares computation agreed with that determined solely on the basis of the 90° and 180° measurements to within one percent.

Representative data from an experiment with a liquid source containing Am^{243} in 3.0M $HClO_4$ are presented in Table IX. The curve representing the angular correlation function obtained from these data by means of a least-squares computation is shown in Fig. 15.

Table IX. Representative Data from an Angular Correlation Experiment with a Liquid Source Containing Am^{243} in Dilute Perchloric Acid

Run No. 37
Activity: Am^{243}; 2.16 μg per λ of solution
Acid concentration: 3.0 molar

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	60	60	60	60	60	60	60
Gross alpha singles ($\times 10^{-2}$)	4551	4549	4548	4544	4543	4542	4549
Alpha background ($\times 10^{-2}$)	24	24	24	24	24	24	24
Net alpha singles ($\times 10^{-2}$)	4527	4525	4524	4520	4519	4518	4525
Gross gamma singles ($\times 10^{-2}$)	4463	4468	4470	4470	4480	4488	4495
Gamma background ($\times 10^{-2}$)	106	106	106	106	106	106	106
Net gamma singles ($\times 10^{-2}$)	4357	4362	4364	4364	4374	4382	4389
Gross triple coincidences ($\times 10^{-1}$)	111	104	97	91	79	75	72
Chance and beta-gamma coincidences ($\times 10^{-1}$)	1	1	1	1	1	1	1
Coincidences due to scattered radiation ($\times 10^{-1}$)	7.8	7.6	7.3	6.9	6.6	6.3	6.2
Net triple coincidences ($\times 10^{-1}$)	102.2	95.4	88.7	83.1	71.4	67.7	64.8
TOTALS							
Number of measurements	8	7	8	8	8	8	8
Total counting time (min)	480	420	480	480	480	480	480
Gross triple coincidences	8750	7380	7870	7090	6510	6130	5960
Spurious triple coincidences	706	606	670	639	609	585	578
Net triple coincidences	8044	6774	7200	6451	5901	5545	5382
Standard deviation (%)	1.3	1.1	1.3	1.0	1.6	1.0	1.2

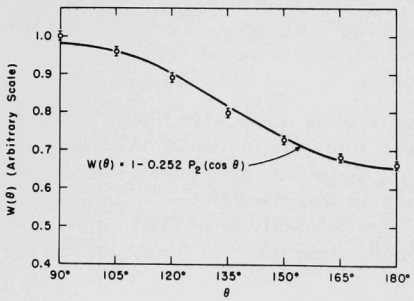

Fig. 15. Experimental Anisotropy for Liquid Source Containing Am^{243} in 3.0 Molar $HClO_4$. The curve was obtained by making a least-squares fit of the data to the function $W(\theta) = 1 + A_2P_2(\cos\theta)$. The correction for detector geometry has not been made.

Results for all of the measurements from liquid sources containing Am^{243} in dilute perchloric acid solutions are summarized in Table X. Data from which least-squares calculations of the angular correlation function could be made were obtained in runs numbered 30, 31, 32, and 37. For the remainder of the runs, measurements were made at 90° and at 180°, and the anisotropy was computed from these measurements. In Table X, standard deviations are indicated for the experimentally determined parameters but not for the derived parameters. All values of A_2 and A have been corrected for detector solid angles and for the isotropic contribution from the triple cascade.

Table X. Results for Liquid Sources Containing Am^{243} in Dilute Perchloric Acid Solutions

Experiment Number	Quantity of Solution	Am^{243} Concentration ($\mu g/\lambda$)	Acid Concentration	A_2	A	G_2
27	2λ	2.37	1M	-0.32	-0.419 ± 0.017	0.90
30	2λ	2.37	1M	-0.335 ± 0.017	-0.43	0.93
31	2λ	2.37	1M	-0.321 ± 0.013	-0.42	0.89
32	2λ	2.37	1M	-0.330 ± 0.006	-0.42	0.92
						ave = 0.91 ± 0.02
33	3λ	2.37	1M	-0.36	-0.458 ± 0.005	1.00
35	3λ	2.37	1M	-0.36	-0.459 ± 0.007	1.00
36	3λ	2.37	1M	-0.35	-0.448 ± 0.007	0.98
						ave = 0.99 ± 0.01
37	3λ	2.16	3M	-0.303 ± 0.013	-0.40	0.84
38	3λ	2.16	3M	-0.32	-0.415 ± 0.006	0.89
39	3λ	2.16	3M	-0.31	-0.398 ± 0.009	0.85
40	3λ	2.16	3M	-0.30	-0.397 ± 0.004	0.85
42	3λ	2.16	3M	-0.31	-0.404 ± 0.005	0.86
						ave = 0.86 ± 0.01
44	3λ	2.14	0.53M	-0.36	-0.458 ± 0.005	1.00
45	3λ	2.14	0.53M	-0.36	-0.452 ± 0.010	0.99
46	3λ	2.14	0.53M	-0.35	-0.451 ± 0.005	0.98
47	3λ	2.14	0.53M	-0.36	-0.454 ± 0.004	0.99
						ave = 0.99 ± 0.01

Initially, liquid sources were prepared with only 2λ of solution. Calculations of the depth of solution from which alpha particles could originate and just reach the plastic scintillator with zero energy after passing through the solution, the rubber hydrochloride membrane, and the helium atmosphere between source and detector showed that, in a 2-λ source, alpha particles from the rear portion of the source could be detected. Recoil nuclei originating near the rear edge of a source can escape from the solution, coming to rest in the glass cover plate used to hold the solution against the rubber hydrochloride membrane. In order to insure that whenever an alpha particle was detected the recoil nucleus would be stopped in the solution, the amount of solution in a source was increased to 3λ. The alpha count rate for the 3-λ sources did not increase over that for the 2-λ sources, a result which indicated that alphas from the rear third of

the 3-λ sources were not detected. However, the anisotropy obtained with the 3-λ sources was significantly greater than that obtained with the 2-λ sources.

C. Even-even Nuclei - Liquid Sources

Angular correlation measurements on the 0-2-0 alpha-gamma transition in even-even nuclei have been made with liquid sources containing either U^{232}, Th^{230}, or Ra^{226} in dilute perchloric acid solutions. Since the theoretical angular correlation function for the 0-2-0 transition is symmetric about 135° and is zero at both 90° and 180°, an anisotropy A could not be determined, and it was necessary to follow the same procedure in obtaining data as was followed for the Am^{243} solid sources. Because of the relatively low gamma activity of the even-even nuclide liquid sources, it was necessary to count these sources for times that were much longer than those used for the Am^{243} sources. Single measurement-counting times were 2 hr for the Th^{230} sources and one hour for the U^{232} and Ra^{226} sources. Complete runs occupied from seven to nine days for the Th^{230} sources and from 74 to 88 hr for the U^{232} and Ra^{226} sources. That the sources remained in the liquid state during the time that it took to complete the runs was determined by periodic observations of the shape of the alpha spectrum.

Representative data from liquid source experiments for each of the even-even nuclides studied are presented in Tables XI-XIV. In spite of the longer counting times used for the even-even nuclide sources, the statistical accuracies obtained with these sources are not as good as those obtained with the Am^{243} sources. Because of the difference in counting rate between Th^{230} sources in 2.2M ClO_4^- and those in 1.1M ClO_4^-, two sets of data are presented for the Th^{230} sources. The angular correlation functions obtained from the data of Tables XI-XIV by means of least-squares computations are shown in Figs. 17-20. The coefficients of the functions shown in Figs. 17-20 have not been corrected for detector solid angle. For purposes of comparison, the correlation function which would be observed for an unperturbed correlation with the detectors used in these measurements is shown in Fig. 16.

It was not necessary to correct the even-even nuclide angular correlation coefficients for the alpha-gamma-gamma triple cascade which originates in alpha decay to the 4+ level in these nuclei. The ratio of alpha decay to the 4+ level to alpha decay to the 2+ level is only 1.0 percent in U^{232}, 0.85 percent in Th^{230}, and 0.2 percent in Ra^{226}.

Results for all of the measurements on liquid sources containing even-even alpha emitters in dilute perchloric acid solutions are summarized in Table XV. All coefficients given in this table have been corrected for the finite size of the detectors. Values for the attenuation coefficients G_2 and G_4 are based on the theoretical correlation function

$$W(\theta) = 1 + 0.714P_2(\cos\theta) - 1.714P_4(\cos\theta) \quad .$$

Table XI. Representative Data from an Angular Correlation Experiment with a Liquid Source Containing Th^{230} in Dilute Perchloric Acid

Run No. 51
Activity: Th^{230}; 98 μg per λ of solution
Acid Concentration: 2.2 molar in perchlorate ion concentration

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	120	120	120	120	120	120	120
Gross alpha singles ($x10^{-3}$)	3647	3632	3656	3652	3625	3642	3602
Alpha background ($x10^{-3}$)	28	28	28	28	28	28	28
Net alpha singles ($x10^{-3}$)	3619	3604	3628	3624	3597	3614	3574
Gross gamma singles ($x10^{-2}$)	780	798	844	847	843	847	857
Gamma background ($x10^{-2}$)	482	507	547	551	549	553	563
Net gamma singles ($x10^{-2}$)	298	291	297	296	294	294	294
Gross triple coincidences	22	36	88	109	88	44	31
Chance coincidences	2	2	2	2	2	2	2
Background coincidences	4	4	4	4	4	4	4
Coincidences due to scattered radiation	3	4	4	5	5	3	3
Net triple coincidences	13	26	78	98	77	35	22
TOTALS							
Number of measurements	13	13	13	13	13	13	13
Total counting time (min)	1560	1560	1560	1560	1560	1560	1560
Gross triple coincidences	327	603	1142	1449	1182	631	367
Spurious triple coincidences	117	130	130	143	143	117	117
Net triple coincidences	210	473	1012	1306	1039	514	250
Standard deviation (%)	8.6	6.9	3.6	2.9	3.1	6.1	10.9

Table XII. Representative Data from an Angular Correlation Experiment with a Liquid Source Containing Th^{230} in Dilute Perchloric Acid

Run No. 55
Activity: Th^{230}; 48 μg per λ of solution
Acid Concentration: 1.1 molar in perchlorate ion concentration

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	120	120	120	120	120	120	120
Gross alpha singles ($x10^{-3}$)	2354	2348	2355	2367	2391	2419	2441
Alpha background ($x10^{-3}$)	1	1	1	1	1	1	1
Net alpha singles ($x10^{-3}$)	2353	2347	2354	2366	2390	2418	2440
Gross gamma singles ($x10^{-2}$)	499	494	536	559	551	552	551
Gamma background ($x10^{-2}$)	381	377	416	438	433	434	435
Net gamma singles ($x10^{-2}$)	118	117	120	121	118	118	116
Gross triple coincidences	15	27	44	68	44	24	20
Chance coincidences	1	1	1	1	1	1	1
Background coincidences	1	1	1	1	1	1	1
Coincidences due to scattered radiation	2	2	2	3	3	1	1
Net triple coincidences	11	23	40	63	39	21	17
TOTALS							
Number of measurements	12	12	12	11	12	12	12
Total counting time (min)	1440	1440	1440	1320	1440	1440	1440
Gross triple coincidences	166	363	672	747	553	289	187
Spurious triple coincidences	48	48	49	55	60	38	41
Net triple coincidences	118	315	623	692	493	251	146
Standard deviation (%)	12.7	6.8	4.1	4.3	5.2	9.3	10.7

Table XIII. Representative Data from an Angular Correlation Experiment with a Liquid Source Containing U^{232} in Dilute Perchloric Acid

Run No. 56
Activity: U^{232}; 0.48 μg per λ of solution
Acid Concentration: 0.53 molar

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	60	60	60	60	60	60	60
Gross alpha singles ($\times 10^{-4}$)	1810	1810	1804	1812	1810	1814	1815
Alpha background ($\times 10^{-4}$)	1	1	1	1	1	1	1
Net alpha singles ($\times 10^{-4}$)	1809	1809	1803	1811	1809	1813	1814
Gross gamma singles ($\times 10^{-2}$)	573	578	588	586	587	583	585
Gamma background ($\times 10^{-2}$)	98	102	110	110	111	111	113
Net gamma singles ($\times 10^{-2}$)	475	476	478	476	476	472	472
Gross triple coincidences	56	157	277	334	280	137	96
Chance coincidences	10	10	10	10	10	10	10
Background coincidences	1	1	1	1	1	1	1
Coincidences due to scattered radiation	9	14	18	19	13	10	7
Net triple coincidences	36	132	248	304	256	116	78
TOTALS							
Number of measurements	10	10	10	10	10	10	10
Total counting time (min)	600	600	600	600	600	600	600
Gross triple coincidences	651	1579	2861	3270	2651	1374	883
Spurious triple coincidences	199	248	287	303	240	207	180
Net triple coincidences	452	1331	2574	2967	2411	1167	703
Standard deviation (%)	6.4	3.3	1.6	1.0	2.4	2.9	4.7

Table XIV. Representative Data from an Angular Correlation Experiment with a Liquid Source Containing Ra^{226} in Dilute Perchloric Acid

Run No. 61
Activity: Ra^{226}; 5.0 μg per λ of solution
Acid Concentration: 0.5 molar

	90°	105°	120°	135°	150°	165°	180°
SINGLE MEASUREMENT							
Counting time (min)	60	60	60	60	60	60	60
Gross alpha singles ($\times 10^{-4}$)	393	393	393	395	395	395	395
Alpha background ($\times 10^{-4}$)	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Net alpha singles ($\times 10^{-4}$)	393	393	393	395	395	395	395
Gross gamma singles ($\times 10^{-2}$)	1974	1969	1972	1950	1949	1944	1960
Gamma background ($\times 10^{-2}$)	240	234	232	234	230	230	220
Net gamma singles ($\times 10^{-2}$)	1734	1735	1740	1716	1719	1714	1740
Gross triple coincidences	78	246	502	613	405	202	128
Chance coincidences	8	8	8	8	8	8	8
Background coincidences	0	0	0	0	0	0	0
Coincidences due to scattered radiation	2	6	17	15	11	7	5
Net triple coincidences	68	232	477	590	386	187	115
TOTALS							
Number of measurements	10	10	10	10	10	10	10
Total counting time (min)	600	600	600	600	600	600	600
Gross triple coincidences	743	2429	5219	6007	4370	2081	1182
Spurious triple coincidences	100	140	250	230	186	150	130
Net triple coincidences	643	2289	4969	5777	4184	1931	1052
Standard deviation (%)	4.0	2.9	1.2	1.6	2.3	2.2	3.3

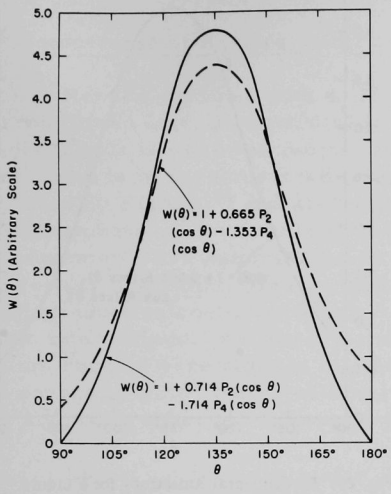

Fig. 16. Theoretical Alpha-Gamma Angular Correlation Functions for the 0-2-0 Transition. The solid curve shows the true theoretical correlation function. The dashed curve shows the correlation function which would be experimentally observed for an unperturbed correlation with the detector geometry used in these measurements.

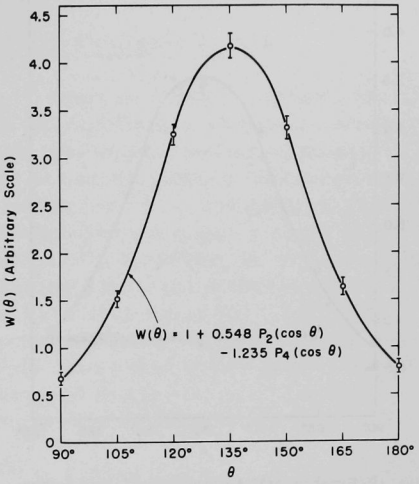

Fig. 17. Experimental Anisotropy for Th^{230} in a Liquid Source Which Was 2.2 Molar in Perchlorate Ion Concentration. The curve was obtained by making a least-squares fit of the data to the function $1 + A_2P_2(\cos\theta) + A_4P_4(\cos\theta)$. The correction for detector geometry has not been made.

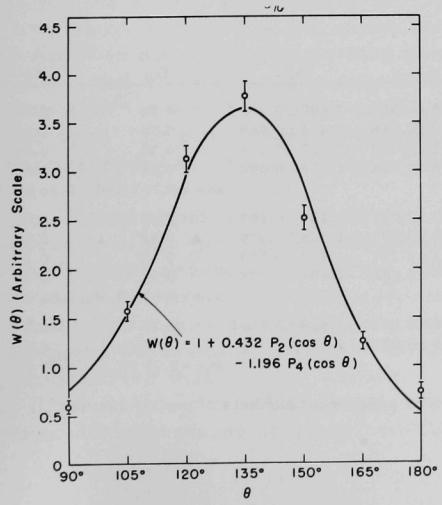

Fig. 18. Experimental Anisotropy for Th^{230} in a Liquid Source Which Was 1.1 Molar in Perchlorate Ion Concentration. The curve was obtained by making a least-squares fit of the data to the function $1 + A_2P_2(\cos\theta) + A_4P_4(\cos\theta)$. The correction for detector geometry has not been made.

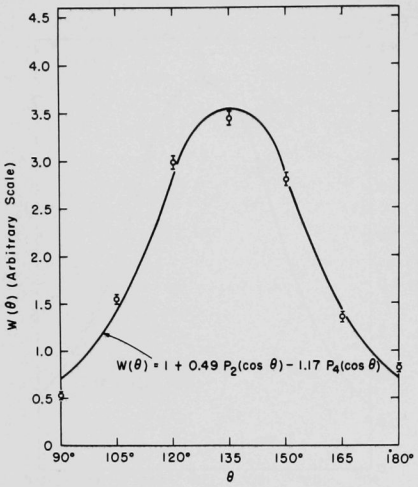

Fig. 19. Experimental Anisotropy for Liquid Source Containing U^{232} in 0.53 Molar $HClO_4$. The curve was obtained by making a least-squares fit of the data to the function $W(\theta) = 1 + A_2P_2(\cos\theta) + A_4P_4(\cos\theta)$. The correction for detector geometry has not been made.

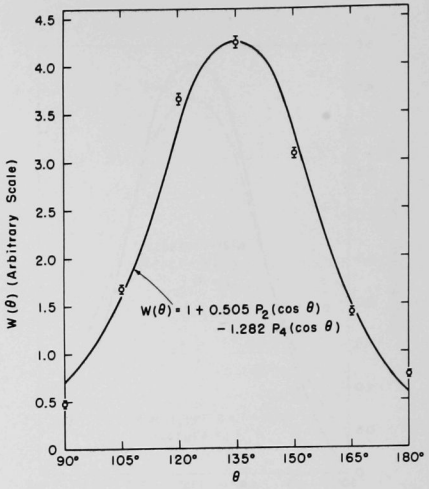

Fig. 20. Experimental Anisotropy for a Liquid Source Containing Ra^{226} in 0.5 Molar $HClO_4$. The curve was obtained by making a least-squares fit of the data to the function $1 + A_2P_2(\cos\theta) + A_4P_4(\cos\theta)$. The correction for detector geometry has not been made.

Table XV. Results for Liquid Sources Containing Even-even Alpha Emitters in Dilute Perchloric Acid Solutions

Experiment Number	Parent Nuclide	Activity Concentration ($\mu g/\lambda$)	Acid Concentration	A_2	A_4	G_2	G_4
56	U^{232}	0.48	0.53M	+0.528 ± 0.060	-1.490 ± 0.095	0.74	0.87
57	U^{232}	0.48	0.53M	+0.539 ± 0.056	-1.482 ± 0.088	0.75	0.86
58	U^{232}	0.45	3.1M	+0.541 ± 0.072	-1.457 ± 0.113	0.76	0.85
59	U^{232}	0.45	3.1M	+0.536 ± 0.058	-1.477 ± 0.092	0.75	0.86
						ave = 0.75 ± 0.05	0.86 ± 0.03
50	Th^{230}	98	0.5M*	+0.485 ± 0.071	-1.404 ± 0.109	0.68	0.82
51	Th^{230}	98	0.5M*	+0.582 ± 0.006	-1.528 ± 0.010	0.82	0.89
54	Th^{230}	48	0.25M*	+0.301 ± 0.101	-1.290 ± 0.155	0.42	0.75
55	Th^{230}	48	0.25M*	+0.460 ± 0.088	-1.481 ± 0.138	0.64	0.86
						ave = 0.64 ± 0.06	0.84 ± 0.04
60	Ra^{226}	4.0	0.5M	+0.540 ± 0.073	-1.529 ± 0.114	0.76	0.89
61	Ra^{226}	4.0	0.5M	+0.538 ± 0.079	-1.570 ± 0.123	0.75	0.92
						ave = 0.76 ± 0.11	0.90 ± 0.07

*The perchlorate ion concentration was 2.2M for experiment numbers 50 and 51 and was 1.1M for experiment numbers 54 and 55.

V. DISCUSSION

A. Comparison of Results with Previously Published Values

Results obtained with solid Am^{243} sources showed that the angular correlation is less attenuated when the Np recoils are stopped in conducting media than it is when the recoils are stopped in nonconducting media. This conclusion is in agreement with results for alpha-gamma measurements reported by Flamm[8] and also with results for numerous gamma-gamma measurements. [The results of the gamma-gamma angular correlation measurements are summarized by Steffen.[3]] However, in contrast with the results of Flamm, present measurements with solid sources did not yield attenuation coefficients below the hard core value ($G_2 = 0.20$) for static interactions. Anisotropies obtained with sources in which the neptunium recoils were stopped in gold were lower than those obtained with the source in which the recoils were stopped in aluminum. This effect has also been observed in gamma-gamma experiments.[3,9]

Liquid sources containing Am^{243} in 0.53 M and 1.0 M $HClO_4$ gave values of the anisotropy in good agreement with the currently accepted theoretical value for the correlation between alpha particles from Am^{243} and the 74.6-kev gamma from Np^{239}. In experiments with liquid sources containing Am^{241} in dilute aqueous solutions, Novey's group[14] obtained a value of $\lambda_2 = 13.5 \times 10^6$ sec^{-1} for the relaxation coefficient associated with exponential decay of the anisotropy in dilute perchloric acid. Since λ_k is a function only of the chemical and physical environment of the source, it is to be expected that the value of λ_2 found by Novey et al. would also be valid for the Am^{243} liquid-source measurements. By using this value of λ_2, a value of $G_2 = 0.98$ is calculated for integral correlation measurements with Am^{243} in dilute perchloric acid solutions. In contrast with the results for 0.5 M and 1.0 M solutions, measurements with liquid sources containing Am^{243} in 3.0 M perchloric acid showed some attenuation of the anisotropy. The attenuation coefficient for these sources was $G_2 = 0.86$. This corresponds to a value of $\lambda_2 = 9.4 \times 10^7$ sec^{-1}.

Measurements with liquid sources containing even-even alpha emitters in dilute perchloric acid solutions also gave attenuated values for the angular correlation function. The attenuation was not, however, as great as has previously been reported for experiments with solid sources. There was generally good agreement between the attenuation coefficients observed with each of the even-even nuclides studied. Variations observed with the Th^{230} sources might have been due in part to the high thorium concentrations used in the sources and in part to the low rates of coincidence counting obtained with these sources.

B. Effect of Source Size on the Measured Value of the Angular Correlation

Correction factors used to correct for the finite size of the detectors employed in angular correlation measurements can be found in the literature, and these correction factors have been applied in the present measurements, as explained in Chapter III. To obtain these correction factors, which assume point sources of radioactivity, the average correlation function $\overline{W(\theta)}$ is determined from the equation

$$\overline{W(\theta)} = \frac{\iint W(\gamma) d\Omega_1 d\Omega_2}{\iint d\Omega_1 d\Omega_2} ,$$

where θ represents the angle between the axes of the two detectors, γ is the angle between the propagation vectors of particles incident on the detectors, and Ω_1 and Ω_2 are the solid angles subtended at the source by the two detectors. For correlations involving gamma radiation, a correction for the efficiency of the detectors as a function of angle must also be included. In principle, a correction for the finite size of the source could be made by extending the above integrations to include the source. Thus, for a finite source,

$$\overline{W(\theta)} = \frac{\iiint W(\gamma) d\sigma d\Omega_1 d\Omega_2}{\iiint d\sigma d\Omega_1 d\Omega_2} ,$$

where $d\sigma$ is an element of surface on the source. Because of the mathematical difficulties involved in actually performing the integration, the correction for an experimental arrangement consisting of finite cylindrical detectors and a finite circular source has not been explicitly carried out. However, an estimate of the correction which should be applied to take into account the finite size of the source can be obtained by considering the case of a finite source and point detectors. For this case,

$$\overline{W(\theta)} = \frac{\int W(\gamma) d\sigma}{\int d\sigma} .$$

Numerical integration of this last expression at the angles $\theta = 90°$ and $\theta = 135°$ was carried out for the experimental arrangement used in these measurements and for the angular correlation function for the 0-2-0 transition in even-even nuclides. Assumptions were made that (1) the correction introduced as a result of the finite extension of the source along the axis of rotation is negligible compared with the correction due to extension of the source perpendicular to the axis of rotation, and (2) the half angle subtended by the source at the center of the gamma detector is negligible compared with the half angle subtended by the source at the center of the alpha detector (these angles are actually 2° 30' and 5° 40', respectively). Error introduced by the second assumption was

partially compensated by considering the source as being perpendicular to the axis of the alpha detector instead of inclined at 45°. The results of the integrations were W(90°) = 0.0199 and W(135°) = 1.856. The correct theoretical values for a point source and point detectors are W(90°) = 0 and W(135°) = 1.874. The integrated values of W(90°) and W(135°) correspond to values of $A_2 = 0.704$ and $A_4 = -1.675$ for the coefficients in the angular correlation function. Thus, it is probable that a correction for the finite size of the source would not change the experimental results by more than 2 or 3 percent.

C. Effect of Nuclear Environment on the Measured Correlation in Solid Sources

The time needed for completion of the recoil motion of the daughter nucleus following alpha decay is of the order of 10^{-13} sec. On the basis that attenuation of the angular correlation will take place during recoil only if the condition $\omega\tau_R > 0.1$ is satisfied, where τ_R is the recoil time, the required interaction frequencies must be greater than 10^{12} sec^{-1}. Since the maximum interaction frequencies so far reported for either electric or magnetic interactions are of the order of 10^9 sec^{-1}, it is extremely unlikely that any attenuation of the correlation takes place during the time of recoil.

Interactions between the emitted alpha particle and the electron shell leave the residual atom following alpha decay in an excited or ionized state. Since an electric field gradient is produced at the nucleus by electrons in incomplete shells whose charge distribution is not spherically symmetric, and since magnetic fields are also produced by atomic electron configurations for which $J \neq 0$, the process of alpha decay gives rise to perturbing electric and magnetic fields within the atom which may interact with sufficient strength to change the spin orientation of the daughter nucleus. The recovery time for an excited or ionized atom in a metal is believed to be of the order of 10^{-12} sec, whereas in a nonconductor the recovery time may be much longer, even of the order of microseconds.[3] This difference in recovery time between a metal and an insulator could be the factor responsible for the greater attenuation observed with solid sources for which the recoils went into a nonconducting medium. Since the ground-state electron configuration for neptunium is probably either $5f^4 6d 7s^2$ or $5f^5 7s^2$,[65] the unpaired 6d or 5f electron could give rise to perturbing fields even after the atom has returned to the ground state.

At the conclusion of its recoil motion, the daughter nucleus from alpha decay is probably located at an interstitial position in the crystal lattice. Furthermore, as a result of Coulomb interactions with the recoiling nucleus, atoms in the crystal may be displaced from their normal positions. Therefore, it is probable that even in crystals, such as aluminum and gold, which normally possess cubic symmetry there will be electric

field gradients at the site where the recoil nucleus comes to rest. Greater lattice distortion in the gold lattice has been postulated as a reason for the increased attenuation observed in gamma-gamma angular correlation experiments with Cd^{111} in a gold lattice as compared with similar experiments carried out in a silver environment.(3) In the present measurements, the angular correlation was found to be more attenuated for sources in which the recoils were stopped in gold than it was for the source in which the recoils were stopped in aluminum.

D. Effect of Liquid Environment on the Angular Correlation

The time required for completion of the nuclear recoil following alpha decay in aqueous solutions is approximately 3.5×10^{-13} sec. Thus, in liquid sources as in solid sources, it is extremely unlikely that there is any appreciable reorientation of the nuclear spin axis during recoil. Furthermore, since the attenuation of the correlation in liquid sources containing Am^{243} was small, it appears that excitation or ionization of the electron shells of the daughter atom is quickly neutralized in the liquid. This is to be expected because of the high collision frequency of ions in solutions.

On the assumption that the measured angular correlation function for liquid sources containing even-even alpha emitters in dilute perchloric acid is perturbed by a time-dependent interaction, the ratio λ_4/λ_2 can be calculated from the observed values of G_2 and G_4 by means of Eq. (10). For the sources containing U^{232}, $\lambda_4/\lambda_2 = 0.49 \pm 0.05$. The value to be expected for this ratio on the basis of a time-dependent electric interaction [see Eq. (11)] is $(\lambda_4/\lambda_2)_e = 0.59$, and the value expected for a time-dependent magnetic interaction [see Eq. (12)] is $(\lambda_4/\lambda_2)_m = 3.3$. The experimentally observed ratio is in good agreement with the postulate of a time-dependent electric interaction, but in complete disagreement with the postulate of a time-dependent magnetic interaction.

On the supposition that the neptunium nucleus is also subject to a time-dependent interaction, one can obtain, using Eq. (11), values for the root-mean-square electric field gradients present in the liquid sources. By using $\lambda_2 = 13.5 \times 10^6$ sec^{-1}, a correlation time τ_c equal to 1×10^{-11} sec,(66,67) and the assumption that the quadrupole moment of the excited neptunium nucleus is not significantly different from the measured moment for the ground-state americium nucleus, one obtains a value of $\left\langle \frac{\partial E_z}{\partial z} \right\rangle = 0.623 \times 10^{18}$ v/cm^2 for the root-mean-square electric field gradient in the liquid sources containing Am^{243} in 0.5 M and 1.0 M $HClO_4$. By using $\lambda_2 = 9.4 \times 10^7$ sec^{-1}, one calculates that the room-mean-square electric field gradient for Am^{243} in 3.0 M $HClO_4$ is $\left\langle \frac{\partial E_z}{\partial z} \right\rangle = 1.64 \times 10^{18}$ v/cm^2. The use of $G_2 = 0.75$ for liquid sources containing even-even nuclei and the assumption that the quadrupole moment for even-even nuclides is of the order

of magnitude $Q = 5.0 \times 10^{-24}$ cm^2 leads to a value of $\left\langle \frac{\partial E_z}{\partial z} \right\rangle = 2.90 \times 10^{18}$ v/cm^2 for these sources. Although the latter two values for the electric field gradient are not unreasonable, it is difficult to understand why they are so much larger than the field gradient in 0.5 M and 1.0 M $HClO_4$. Alternatively, if one assumes that the nuclear quadrupole moment and the mean-square electric field gradient at the nucleus are approximately the same for all of the nuclides in dilute aqueous solution, Eq. (11) can be used to determine the differences in correlation time which must be postulated to account for the different values of λ_2. The correlation times τ_c for the sources containing Am^{243} in 3.0 M $HClO_4$ must then be 7.0 times as great as the correlation times for the 0.5 M and 1.0 M solutions. The correlation times for sources containing even-even nuclides must be approximately 22 times as great as those of the sources containing Am^{243} in 0.5 M and 1.0 M $HClO_4$.

These differences are all the more puzzling if one considers the most stable charge states of the daughter nuclei in dilute acid solution. The normal oxidation states for thorium and radium in solution are Th^{+4} and Ra^{+2}, respectively. In these oxidation states, the thorium would have eight oxygens coordinated to it (i.e., eight waters of hydration), and the radium would have four oxygens coordinated to it. These coordination spheres should produce spherically symmetric charge distributions about the thorium and radium nuclei. The radon daughter which follows alpha decay of Ra^{226} is completely inert in the ground state and should not even be surrounded by a coordination sphere of water molecules. Hence, any field gradients present at the site of the radon nucleus should be only those normally present in dilute acid solutions.

With neptunium, the ions Np^{+3}, Np^{+4}, NpO_2^+, and NpO_2^{+2} are all reasonably stable in dilute acid. The ions Np^{+3} and Np^{+4} should be surrounded by coordination spheres of water molecules in the same manner as are Th^{+4} and Ra^{+2}. The neptunyl ion, NpO_2^{+2}, is pictured as having a linear structure, O-Np-O, in which the two oxygens are very strongly bonded to the neptunium. Such a structure would give rise to an appreciable field gradient along the bond axis of the molecule. However, in solution the neptunyl ion is surrounded by six water molecules which form a puckered ring about the equator of the ion.[68,69] This arrangement could result in a low value of the root-mean-square electric field gradient at the neptunium nucleus if the combination of the coordination sphere of water molecules about the equator of the ion together with the two permanently bonded oxygens should give rise to an approximately spherically symmetric charge distribution. Similar considerations probably also apply to NpO_2^+ in dilute acid solutions. Experiments[70,71] have shown that the initial charge on recoil atoms following alpha decay is one or two positive units. Thus, it might be questioned whether the neptunium and the thorium would reach the high charge states postulated above in a time which is short compared with the excited state lifetimes for these nuclei. Transitions to higher charge states could be

accomplished as a result of interactions with free radicals produced in the solutions by the recoiling nuclei. On the other hand, the conditions existing in solution a millimicrosecond after alpha decay cannot be definitely established, and it may well be that the actual oxidation states of the daughter nuclei are not the states discussed here.

One can postulate that, since the theoretical angular correlation function for Am^{243} is sensitive to the mixing ratio of the L = 0 and L = 2 alpha waves and since this mixing ratio is only approximately known, the theoretical correlation function may be in error. The assumption that the correlation for the liquid sources containing Am^{243} in 0.5 M and 1.0 M $HClO_4$ was attenuated by the same amount as the correlation for the liquid sources containing even-even nuclides requires a value of $A_2 = -0.48$ for the americium correlation coefficient. The mixing ratio necessary to give this value of A_2 is approximately $\delta^2 = 0.67$. In the collective model of Bohr and Mottelson the mixing ratio is determined by Eq. (6), in which c_2 is the average of the reciprocals of the L = 2 hindrance factors for neighboring even-even nuclei. If, instead of the average, one takes the largest value of c_2 consistent with neighboring even-even data (i.e., $c_2 = 0.715$; the reciprocal of the hindrance factor of Pu^{242}) a value of $\delta^2 = 0.26$ is obtained for the mixing ratio. This corresponds to a value of $A_2 = -0.38$, which is only 6 percent greater than the value computed from the average hindrance factors.

An alternative postulate is that the 74.6-kev gamma which depopulates the 5/2- state in Np^{239} is a mixture of E1 and M2 radiations. An admixture of one percent of M2 radiation in this transition would give a value of $A_2 = -0.44$ for the coefficient of the Am^{243} angular correlation function. (This value of A_2 assumes that the gamma waves are in phase.) The amount of M2 radiation expected in this transition on the basis of comparisons of experimental and theoretical values of L-shell internal conversion coefficients can be determined from the equation

$$\alpha_L = K\,\alpha_L(E1) + (1 - K)\,\alpha_L(M2) \quad , \tag{15}$$

where

$$K = \frac{\lambda_\gamma(E1)}{\lambda_\gamma(E1) + \lambda_\gamma(M2)} \quad \text{and} \quad 1 - K = \frac{\lambda_\gamma(M2)}{\lambda_\gamma(E1) + \lambda_\gamma(M2)} \quad .$$

Here, $\alpha_L(E1)$ is the L-shell conversion coefficient for E1 radiation and $\lambda_\gamma(E1)$ is the probability per unit time of emission of an E1 gamma photon in the transition. Limits on the error to be expected in the experimentally determined value of the total L-shell conversion coefficient for the 74.6-kev transition in Np^{239} have been set by Asaro et al.[72] as $\alpha_L = 0.20 \pm 0.05$. Theoretical values for L-shell conversion coefficients have been calculated by Rose[73] and by Sliv and Band.[74] By using $\alpha_L = 0.25$ and the theoretical

conversion coefficients of Rose in Eq. (15), one calculates that the maximum contribution of M2 radiation to the 74.6-kev transition in Np^{239} is 0.032%. The use of the theoretical conversion coefficients of Sliv and Band leads to a value of 0.028% for the maximum contribution of M2 radiation. Thus, the postulate that the angular correlation observed for liquid sources containing Am^{243} in 0.5 M and 1.0 M $HClO_4$ is appreciably attenuated seems to be untenable.

E. Conclusions

Measurements with liquid sources containing Am^{243} in dilute perchloric acid solutions showed that the unattenuated correlation was obtained for liquid sources in which the acid concentration was 0.5 M and 1.0 M. For liquid sources in which the acid concentration was 3.0 M, the correlation was attenuated. Measurements with liquid sources containing either U^{232}, Th^{230}, or Ra^{226} in dilute perchloric acid solutions also gave attenuated values of the angular correlation. The attenuation coefficients obtained for the even-even nuclide sources are believed to be consistent with the postulate of a time-dependent interaction between the nuclear quadrupole moment and randomly changing electric field gradients in the liquid. However, in order to explain the liquid results on the basis of a time-dependent electric interaction, it is necessary to assume that either large differences exist between the electric field gradients present in the americium liquid sources and the even-even nuclide liquid sources, or else that large differences exist in correlation times. The processes which take place in dilute aqueous solutions on a millimicrosecond time scale, and the effect of the nuclear recoil on these processes are not well enough understood to make possible a detailed explanation of the difference in nuclear environments which apparently exists in these sources following alpha decay.

Two of the most important uses of an alpha-gamma angular correlation measurement are to provide information about spin assignments of intermediate nuclear levels and to provide information about the relative intensities and phases of the alpha waves which participate in the transition being investigated. In many instances, liquid-source measurements should make it possible to decide among several alternative spin sequences even though the correlation is somewhat perturbed. Measurements with liquid sources should provide unambiguous information about the relative phases of the participating alpha waves. However, because the correlation may be attenuated, such measurements can only be used to set limits on the relative intensities of the alpha waves.

Some useful information about the interactions responsible for attenuation of the alpha-gamma angular correlation in liquid sources might be obtained by using sources containing even-even plutonium or curium isotopes. Such experiments might, for example, be performed with Pu^{238} or Cm^{242}, for

which coincidence rates comparable with those found for Th^{230} sources could be achieved. The results of such experiments could be compared directly with results presented here for even-even nuclides. Uranium and plutonium form the same range of stable oxidation states as does neptunium. If very little attenuation of the correlation were obtained, as with Am^{243}, this would be good evidence that the higher oxidation states MO_2^+ and MO_2^{+2} are formed and that this is the reason for the difference between the results obtained with Am^{243} and the even-even nuclei of lower atomic number. In the event of an attenuated correlation, a choice between an electric or a magnetic interaction as the mechanism responsible for the attenuation could be made from the ratio of G_2 to G_4.

APPENDIX I

Recoil of Daughter Nuclei Following Alpha Decay

For the observation of an unperturbed alpha-gamma angular correlation, it is necessary that the spin of the intermediate nuclear level remain fixed in direction during the intermediate-level lifetime. In alpha decay, an appreciable fraction of the decay energy occurs as recoil energy of the daughter nucleus. As a result of this recoil, the nucleus may be subjected to magnetic fields or to electric field gradients of sufficient magnitude to perturb the spin direction of the intermediate nuclear level. It is therefore important to have some knowledge of the magnitude of the recoil and of the time required for its completion.

From conservation of momentum it can be shown that the neptunium daughter which follows alpha decay of Am^{243} has a recoil energy of 88.3 kev. This corresponds to an initial recoil velocity of 2.83×10^7 cm/sec for the neptunium nucleus.

Nielsen[75] has calculated the range (really the total track length of the recoil ion) for atomic particles with energies of about 50 kev on the assumption that the mass of the moving ion is greater than that of the target atoms and that, for the velocities considered, electron-stopping effects (i.e., ionization and excitation of the target atoms) may be disregarded. The stopping mechanism is considered to be elastic nuclear collisons between the moving ion and target atoms in the stopping material. The range is given by

$$t = 0.6 \frac{(Z_1^{2/3} + Z_2^{2/3})^{1/2}}{Z_1 Z_2} \frac{A_1 + A_2}{A_1} A_2 E_1 \ \mu g/cm^2 \quad ,$$

where the subscript 1 refers to the incoming particles, the subscript 2 refers to the target particles, and E_1 is measured in kev.

A summary of some experimental results reported in the literature for the recoil range of heavy ions in solid materials is presented in Table XVI. The theoretical range calculated from Nielsen's formula is also given. In all cases the calculated range and the experimentally observed mean range agree within a factor of two. It should be noted that Cohen's results showed that there was considerable range straggling and that the maximum range exceeded the mean range by as much as a factor of three.

Calculated ranges and stopping times for 88.3-kev Np^{239} recoil ions in aluminum, gold, and Am_2O_3 are given in Table XVII. It is probable that the calculated ranges are accurate to within a factor of two. Stopping times were calculated on the assumption that the average recoil velocity is one-half the initial recoil velocity.

Table XVI. Comparison of Experimental Results on the Recoil Range of Heavy Ions in Solid Materials with the Range Calculated from Nielsen's Formula

Reference	Recoil Nucleus	Recoil Energy (kev)	Stopping Material	Experimental Range (μg/cm^2) Mean	Experimental Range (μg/cm^2) Maximum	Theoretical Range (μg/cm^2)
76	Tl^{208}	116	gold	34	~100	25.4
76	Tl^{208}	116	silver	12	~ 35	16.9
76	Tl^{208}	116	aluminum	7	~ 16	9.9
76	Tl^{208}	116	collodion	11.6	~ 23	
77	Na^{24}	30	aluminum	13.2		23.4
77	Rb^{30}	30	aluminum	6.6		5.4
77	Cs^{137}	20	aluminum	4.0		2.4
77	Cs^{137}	30	aluminum	5.7		3.6
77	Cs^{137}	50	aluminum	9.5		6.0
78	Ra^{224}	96.8	He	6.6		6.2
78	Ra^{224}	96.8	Ne	7.3		7.1
78	Ra^{224}	96.8	A	9.4		9.0
78	Po^{208}	140.2	Bi^{209}	23.5		31.2
78	Po^{208}	160.7	Bi^{209}	25.0		35.8
78	Po^{208}	220.4	Bi^{209}	29.2		49.1

Table XVII. Calculated Ranges and Stopping Times of 88.3-kev Np^{239} Recoils in Various Materials

Material	Range (μg/cm^2)	Range (Angstroms)	Stopping Time (sec)
Aluminum	6.7	248	1.75×10^{-13}
Gold	16.2	84	5.92×10^{-14}
Am_2O_3	11.8	~100	$\sim 7 \times 10^{-14}$
Water	5.04	504	3.56×10^{-13}

APPENDIX II

Chemical Purification of Radionuclides

A. Am^{243}

Americium was adsorbed from a 6M HCl solution onto the top of a Dowex 50 x 5 cation resin* column, of 2-mm id by 150 mm long. The column was first washed with 0.5M HCl to remove mono- and di-valent elements, and then eluted with 6M HCl to remove the americium. The americium was fumed to dryness with 70% $HClO_4$ and redissolved in $HClO_4$ of the desired concentration. The americium concentration varied from 2.1 to 2.4 mg/ml. An alpha pulse analysis showed that 93.4% of the alpha activity was from Am^{243}, 0.88% from Am^{241}, 5.7% from Cm^{244}, and 0.02% from Cm^{242}. The Cm^{244} would cause no interference in the coincidence experiments since Cm^{244} emits only 0.035% as many gammas per alpha as does Am^{243}.

B. Th^{230}

No radiochemical purification was performed since an alpha pulse analysis showed that 99.91 ± 0.02% of the alphas were those from Th^{230}, and a gamma spectrum showed no evidence for any activity other than Th^{230}. The thorium nitrate starting material was converted to the perchlorate by fuming to dryness with 70% $HClO_4$. The dry salt was dissolved in 0.57M $HClO_4$ to give a solution which was 0.5M in $HClO_4$ and 2.2M in total ClO_4^- on the assumption that the salt formed by evaporating to dryness with $HClO_4$ was $Th(ClO_4)_4$. The thorium concentration was 97 mg/ml. This solution was later diluted twofold with water for the experiments at half this concentration.

C. U^{232}

Uranium was adsorbed from a concentrated HCl solution onto the top of a Dowex 1 x 10 anion resin** column, 2 mm in id by 25 mm long. Th^{228} and its daughters were washed from the column with concentrated HCl, and the purified uranium eluted with 6M HCl. The uranium was converted to the perchlorate by fuming to dryness with 70% $HClO_4$ and was then dissolved in 0.5 or 3M $HClO_4$ to give the final solution. An alpha pulse analysis gave a U^{232}/U^{233} alpha activity ratio of 62/1, with no evidence for any other alpha emitters. The total uranium concentration was 16 mg/ml.

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*A copolymer of 95 percent styrene and 5 percent divinylbenzene with sulfonic acid functional groups.

**A copolymer of 90 percent styrene and 10 percent divinylbenzene with trimethylbenzylammonium functional groups.

D. Ra^{226}

A commercial source of $RaBr_2$ with a stated purity of greater than 99% was further purified by four precipitations as $RaCl_2$ from 7M HCl. This procedure effectively removed all daughters. The radium was converted to the perchlorate by fuming to dryness with 70% $HClO_4$ and was then dissolved in 0.5M $HClO_4$. The radium concentration was approximately 4 mg/ml.

APPENDIX III

The Theoretical Angular Correlation Function for Alpha Decay of Am^{243}

Alpha decay of Am^{243} to the 74.6-kev level in Np^{239} proceeds by means of a mixture of L = 0 and L = 2 multipoles. Because the alpha-gamma angular correlation function associated with this transition cannot be calculated from formulae given in the review article by Frauenfelder,[1] details of the calculation using the formalism of Biedenharn and Rose[6]* are presented here.

The angular correlation function for the cascade $j_1(L_1L_1')j(L_2)j_2$ in which the first transition is mixed and consists of the superposition of two angular momenta L_1 and L_1' can be written

$$W(\theta) = W_I(\theta) + W_{II}(\theta) + W_{III}(\theta) \quad .$$

Expressions for W_I, W_{II}, and W_{III} for the case of a gamma-gamma transition are given by BR, Eq. 41a-41c. Here, W_I and W_{II} represent appropriately weighted contributions to $W(\theta)$ from transitions involving pure 2^{L_1} pole and pure $2^{L_1'}$ pole radiations, and W_{III} contains the effect on $W(\theta)$ of the phase difference between the radiations. The intensity ratio δ^2 of $2^{L_1'}$ pole radiation to 2^{L_1} pole radiation is given in terms of reduced matrix elements by BR, Eq. 67.

The equations for a gamma-gamma cascade can be adapted to an alpha-gamma cascade involving a mixture of alpha multipoles by making use of the so-called particle parameter which is defined by BR, Eq. 42. For an alpha transition involving only pure 2^L alpha waves, the particle parameter (BR, Eq. 79) is given by

$$b_n(LL;\ \alpha) = \frac{2L(L+1)}{2L(L+1) - n(n+1)} \quad .$$

For the 5/2(02)5/2(1)5/2 alpha-gamma transition in the alpha decay of Am^{243}, W_I and W_{II} can be written (BR, Eq. 69a, 70, 70a, 70b) in the form

$$W_I = 1 + b_2(L_1L_1;\ \alpha)\ A_2(L_1L_2)P_2(\cos\theta) \quad ;$$

$$W_{II} = \delta^2[1 + b_2(L_1'L_1';\ \alpha)\ A_2(L_1'L_2)P_2(\cos\theta)] \quad ,$$

where

$$A_2(L_1L_2) = F_2(L_1L_1j_1j)F_2(L_2L_2j_2j) \quad .$$

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* In what follows, the paper of Biedenharn and Rose will be referred to as BR.

The F coefficients are defined by BR, Eq. 69b. A short table of F coefficients is given by Biedenharn and Rose, and a more complete tabulation is given in a report by Ferentz and Rosenzweig.[39] Both $b_2(L_1L_1; \alpha)$ and $A_2(L_1L_2)$ are equal to zero (physically, this results from the fact that L = 0 alpha waves have spherical symmetry). By making use of BR, Eq. 78a, W_{III} can be written as

$$W_{III} = -2\delta(2j+1)\,(2L_2+1)[(2L_1+1)(2L_1'+1)]^{1/2}$$
$$\times\, C(L_2L_2 2;\ 1\text{-}1)\ W(jjL_2L_2;\ 2j_2)$$
$$\times\, C(L_1L_1'\,2;\ 0\ 0)\ W(jjL_1L_1';\ 2j_1)$$
$$\times \cos(\sigma_{L_1'} - \sigma_{L_1}) P_2(\cos\theta) \quad .$$

This can be further simplified to

$$W_{III} = -2\delta F_2(L_2L_2j_2j)[(2j+1)(2L_1+1)(2L_1'+1)]^{1/2} C(L_1L_1'2;\ 0\ 0)$$
$$\times\, W(jjL_1L_1';\ 2j_1)\cos(\sigma_{L_1'} - \sigma_{L_1})\, P_2(\cos\theta) \quad .$$

The factor (2j+1) is a normalizing factor which makes A_0 in W_I and W_{II} equal to unity. Tables of Clebsch-Gordan coefficients have been prepared by Simon,[79] and tables of Racah coefficients have been prepared by Simon et al.[80] The quantity σ_L is the phase shift due to the Coulomb potential. For alpha-particle energies well below the height of the Coulomb barrier, Seed and French [81] have shown that the phase difference can be written

$$\sigma_{L+2} - \sigma_L = \tan^{-1}[n/(L+2)] + \tan^{-1}[n/(L+1)] \quad .$$

Here $n \approx 0.63Z/E_\alpha^{1/2}$, where Z is the atomic number of the daughter nucleus and E_α is the alpha energy in Mev.

By combining the above equations for W_I, W_{II}, and W_{III}, the alpha-gamma angular correlation function for the 5/2(02)5/2(1)5/2 transition in the alpha decay of Am^{243} can be written as $W(\theta) = 1 + A_2P_2(\cos\theta)$, where

$$A_2 = \frac{-0.1634\delta^2 - 0.856\delta\cos(\sigma_2 - \sigma_0)}{1 + \delta^2} \quad .$$

For $\delta^2 = 0.22$ and the alpha waves in phase, $A_2 = -0.36$. (For the alpha waves out of phase, $A_2 = +0.30$.)

BIBLIOGRAPHY

1. Frauenfelder, H., Beta- and Gamma-ray Spectroscopy, edited by K. Siegbahn (Interscience Publishers Inc., New York, 1955), p. 531.
2. Devons, S., and L. J. B. Goldfarb, Handbuch der Physik, edited by S. Flugge (Springer-Verlag, Berlin, 1956), Vol. 42, p. 362.
3. Steffen, R. M., Adv. Phys. (Phil. Mag. Suppl.) 4, 293 (1955).
4. Heer, E., and T. B. Novey, Solid State Physics, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1959), Vol 9, p. 199.
5. Hamilton, D. R., Phys. Rev., 58, 122 (1940).
6. Biedenharn, L. C., and M. E. Rose, Revs. Mod. Phys., 25, 729 (1953).
7. Migdal, A., J. Phys. (U.S.S.R.), 4, 499 (1941).
8. Flamm, E., UCRL-9325 (1960).
9. Aeppli, H., A. S. Bishop, H. Frauenfelder, M. Walter, and W. Zunti, Phys. Rev., 82, 550 (1951).
10. Steffen, R. M., Phys. Rev., 90, 1119 (1953).
11. Kraushaar, J. J., and R. V. Pound, Phys. Rev., 92, 522 (1953).
12. Paul, H., and R. M. Steffen, quoted in R. M. Steffen, Adv. Phys. (Phil. Mag. Suppl.), 4, 293 (1955).
13. Novey, T. B., Phys. Rev., 96, 547 (1954).
14. Krohn, V. E., T. B. Novey, and S. Raboy, Phys. Rev., 105, 234 (1957).
15. Stephens, F., J. Hummel, F. Asaro, and I. Perlman, Phys. Rev., 98, 261 (1955).
16. Hummel, J. P., UCRL-3456 (1956).
17. Strominger, D., J. M. Hollander, and G. T. Seaborg, Revs. Mod. Phys., 30, 585 (1958).
18. Unik, J. P., UCRL-9105 (1960).
19. Asaro, F., and I. Perlman, Phys. Rev., 93, 1423 (1954).
20. Conway, J. G., and R. D. McLaughlin, Phys. Rev., 94, 498 (1954).
21. Hubbs, J. C., and R. Marrus, Phys. Rev., 110, 287 (1958).
22. Bohr, A., and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd., 27, No. 16 (1953).
23. Bohr, A., and B. R. Mottelson, Beta- and Gamma-ray Spectroscopy, edited by K. Siegbahn (Interscience Publishers Inc., New York, 1955), p. 468.

24. Bohr, A., Rotational States of Atomic Nuclei (Ejnar Munksgaards Forlag, Copenhagen, 1954).

25. Perlman, I., and J. O. Rasmussen, Handbuch der Physik, edited by S. Flugge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 109.

26. Stephens, F. S., Jr., F. Asaro, and I. Perlman, Phys. Rev., 100, 1543 (1955).

27. Stephens, F. S., Jr., F. Asaro, and I. Perlman, Phys. Rev., 107, 1091 (1957).

28. Asaro, F., and I. Perlman, Phys. Rev., 99, 37 (1955).

29. Scharff-Goldhaber, G., E. der Mateosian, G. Harbottle, and M. McKeown, Phys. Rev., 99, 180 (1955).

30. Asaro, F., and I. Perlman, Phys. Rev., 88, 129 (1952).

31. Harbottle, G., M. McKeown, and G. Scharff-Goldhaber, Phys. Rev., 103, 1776 (1956).

32. Bell, R. E., S. Bjornholm, and J. C. Severiens, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 32, No. 12 (1960).

33. Vartapetian, H., and R. Foucher, Compt. rend., 246, 939 (1958).

34. Evans, R. D., The Atomic Nucleus (McGraw-Hill Book Company, Inc., New York, 1955), p. 155.

35. Perlman, I., and F. Asaro, Ann. Rev. Nuclear Sci., 4, 157 (1954).

36. Rosenblum, S., and M. Valadares, Compt. rend. 234, 2359 (1952).

37. Booth, E., L. Madansky, and F. Rasetti, Phys. Rev., 102, 800 (1956).

38. Roy, R. R., and M. L. Goes, Compt. rend., 238, 469 (1954).

39. Ferentz, M., and N. Rosenzweig, ANL-5324 (1953).

40. Milton, J. C. D., and J. S. Fraser, Phys. Rev., 95, 628 (1954).

41. Valladas, G., J. Teillac, P. Falk-Vairant, and P. Benoist, J. phys. radium, 16, 125 (1955).

42. Temmer, G. M., and J. M. Wyckoff, Phys. Rev., 92, 913 (1953).

43. Battey, J., L. Madansky, and F. Rasetti, Phys. Rev., 89, 182 (1953).

44. Belling, J. K., B. T. Feld, and I. Halpern, Phys. Rev., 84, 155 (1951).

45. Fraser, J. S., and J. C. D. Milton, Phys. Rev., 94, 795 (1954).

46. Petit, G. Y., Compt. rend., 245, 1916 (1957).

47. Horton, J. W., Phys. Rev., 101, 717 (1956).

48. Goertzel, G., Phys. Rev., 70, 897 (1946).

49. Alder, K., Helv. Phys. Acta, 25, 235 (1952).

50. Abragam, A., and R. V. Pound, Phys. Rev., 89, 1306 (1953).

51. Pound, R. V., and A. Abragam, Phys. Rev., 90, 993 (1953).

52. Abragam, A., and R. V. Pound, Phys. Rev., 92, 943 (1953).

53. Kopfermann, H., Nuclear Moments (Academic Press Inc., New York, 1958), p. 132.

54. Manning, T. E., M. Fred, and F. S. Tomkins, Phys. Rev., 102, 1108 (1956).

55. Pake, G. E., Am. J. Phys., 18, 438 (1950).

56. Smith, K. F., Progress in Nuclear Physics, edited by O. R. Frisch (Pergamon Press, New York, 1957), Vol. 6, pp. 81-87.

57. Kopfermann, H., Nuclear Moments (Academic Press Inc., New York, 1958), p. 335.

58. Simmons, W. W., and C. P. Slichter, TID-11051 (1960).

59. Alder, K., A. Bohr, T. Huus, B. Mottelson, and A. Winther, Revs. Mod. Phys., 28, 432 (1956).

60. Kerman, A. K., Nuclear Reactions, edited by P. M. Endt and M. Demeur (Interscience Publishers Inc., New York, 1959), p. 439.

61. Kaplan, I., Nuclear Physics (Addison-Wesley Publishing Company, Inc., Cambridge, Mass., 1955), p. 44.

62. Rose, M. E., Phys. Rev., 91, 610 (1953).

63. Stanford, A. L., Jr., and W. K. Rivers, Jr., Rev. Sci. Instr., 30, 719 (1959).

64. West, H. I., Jr., UCRL-5451 (1959).

65. Hyde, E. K., and G. T. Seaborg, Handbuch der Physik, edited by S. Flugge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 205.

66. Debye, P., Polar Molecules (Dover Publications, New York, 1945), p. 85.

67. Bloembergen, N., E. M. Purcell, and R. V. Pound, Phys. Rev., 73, 679 (1948).

68. Connick, R. E., and Z. Z. Hugus, J. Amer. Chem. Soc., 74, 6012 (1952).

69. Eisenstein, J. C., and M. H. L. Pryce, Proc. Roy. Soc. (London), A229, 20 (1955).

70. Mund, W., P. Capron, and J. Jodogne, Bulletin Soc. Chim. Belgique, 40, 35 (1931).

71. McGee, J. D., Phil. Mag., 13, 1 (1932).

72. Asaro, F., F. S. Stephens, J. M. Hollander, and I. Perlman, Phys. Rev., 117, 492 (1960).

73. Rose, M. E., Internal Conversion Coefficients (Interscience Publishers Inc., New York, 1958).

74. Sliv, L. A., and I. M. Band, Coefficients of Internal Conversion of Gamma Radiation, Part II, L-shell, Report No. 58ICCL1, Physics Department, University of Illinois, Urbana, Illinois (1958).

75. Nielsen, K. O., Electromagnetically Enriched Isotopes and Mass Spectrometry, edited by M. L. Smith (Academic Press Inc., New York, 1956), p. 68.

76. Cohen, S. G., Proceedings of Rehovoth Conference on Nuclear Structure, (North-Holland Publishing Co., Amsterdam, 1958), p. 580.

77. Davies, J. A., Abstracts of Papers Presented at the Third Symposium on Nuclear and Radiochemistry, Chalk River, Ontario, Canada, Sept. 6-8, 1960, Paper No. 2.

78. Valyocsik, E. W., UCRL-8855 (1959).

79. Simon, A., ORNL-1718 (1954).

80. Simon, A., J. H. Vander Sluis, and L. C. Biedenharn, ORNL-1679 (1954).

81. Seed, J., and A. P. French, Phys. Rev., 88, 1007 (1952).

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